SHORT NOTES

Resonance spin modes in layered conductors

D. I. Stepanenko*

B. Verkin Institute for Low Temperature Physics and Engineering, National Academy of Sciences of Ukraine, pr. Lenina 47, Kharkov 61103, Ukraine
(Submitted May 6, 2004)
Fiz. Nizk. Temp. **31**, 115–119 (January 2005)

The propagation of spin waves in layered conductors in the presence of an external magnetic field is studied theoretically. It is it is shown that for certain orientations of the magnetic field with respect to the layers of the conductor there is no collisionless absorption, and weakly damped collective modes can propagate even under conditions of strong spatial dispersion. © *2005 American Institute of Physics.* [DOI: 10.1063/1.1820379]

In recent years there has been a significant increase in interest in layered structures with a metallic type of conductivity and a quasi-two-dimensional electron energy spectrum. These include organic conductors of the family of tetrathiafulvalene salts, transition metal dichalcogenides, graphite, etc. In the absence of external magnetic field the conductivity of these substances along the layers, $\sigma_\parallel$, is several orders of magnitude greater than the conductivity across the layers, $\sigma_\perp$. The marked anisotropy of the kinetic coefficients of layered conductors is a consequence of the quasi-two-dimensionality of their electron energy spectrum. The maximum velocity of electrons with the Fermi energy ε_F along the normal $\mathbf{n}$ to the layers, $v_z=\mathbf{v}\cdot\mathbf{n}$, is much less than the characteristic velocity of electrons in the plane of the layers, v_F, and their energy can be written in the form of a rapidly converging series:

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

The functions $\varepsilon_n(p_x,p_y,\eta)$ fall off substantially with increasing index:

$$\varepsilon_{n+1}(p_x,p_y,\eta)\ll\varepsilon_n(p_x,p_y,\eta),\varepsilon_1(p_x,p_y,\eta)\sim\eta\varepsilon_F.$$

Here $\eta=(\sigma_\perp/\sigma_\parallel)^{1/2}$ is the quasi-two-dimensionality parameter, $p_0=\hbar/a$, $\hbar$ is Planck's constant, and a is the distance between layers. Formula (1) corresponds to the tight binding approximation, when the overlap of the electron shells of atoms belonging to different layers is small and the distance between layers is much greater than the interatomic distance within a layer. The Fermi surface $\varepsilon(\mathbf{p})=\varepsilon_F$ corresponding to dispersion relation (1) is open, with a slight corrugation along the p_z axis; it can be multisheet and consist of topologically different elements, e.g., cylinders and planes. It will be assumed from here on that the Fermi surface of the layered conductor is a slightly corrugated cylinder, all sections of which by the plane $p_B=(\mathbf{p}\cdot\mathbf{B}_0)/B_0=p_z\cos\vartheta+p_x\sin\vartheta=\text{const}$ are closed for $\pi/2-\vartheta>\eta$; here $\mathbf{B}_0=(B_0\sin\vartheta,0,B_0\cos\vartheta)$ is the external magnetic induction. Numerous experimental studies of magnetic oscillations have shown that such a Fermi surface is possessed by a considerable number of organic conductors based on tetrathiafulvalene salts.[1]

In normal metals at low temperatures in a magnetic field, various weakly damped collective modes of the Bose type can exist: electromagnetic, sound, and spin waves. In layered conductors the propagation of collective modes differs in a number of features due to the topology of the Fermi surface. For certain orientations of the magnetic field relative to the layers of the conductor the projection of the electron velocity on the direction of $\mathbf{B}_0$, averaged over the period of motion of an electron along the cyclotron orbit, is negligible. For those directions of $\mathbf{B}_0$ collisionless absorption is absent, and the propagation of weakly damped waves is possible even under conditions of strong spatial dispersion. In this communication we report an investigation of spin waves in layered conductors with a quasi-two-dimensional electron energy spectrum. Collective modes involving oscillations of the spin density of the conduction electrons in quasi-isotropic conductors lacking magnetic order were predicted by Silin[2] and observed experimentally in alkali metals by Schultz and Dunifer.[3]

In the case when the condition $\hbar\omega_B\lesssim T\ll\eta\varepsilon_F$ is met (where T is the temperature and ω_B is the cyclotron frequency of a conduction electron), the density matrix $\hat{\rho}$ is an operator in the space of spin variables and a quasiclassical function of the coordinates and momenta, while the additional energy of the quasiparticle due to electron–electron interaction effects can be written in the framework of the Landau–Silin theory of the Fermi liquid:

$$\delta\hat{\varepsilon}(\mathbf{p},\mathbf{r},t)=\mathrm{Tr}_{\sigma'}\int\frac{d^3p'}{(2\pi\hbar)^3}L(\mathbf{p},\hat{\sigma},\mathbf{p}',\hat{\sigma}')\,\delta\rho'(\mathbf{p}',\mathbf{r},\hat{\sigma}',t), \quad (2)$$

where $L(\mathbf{p},\hat{\sigma},\mathbf{p}',\hat{\sigma}')=N(\mathbf{p},\mathbf{p}')+S(\mathbf{p},\mathbf{p}')\hat{\sigma}\hat{\sigma}'$ is the Landau correlation function, $\delta\hat{\rho}$ is the nonequilibrium admixture to the density matrix, and $\hat{\sigma}$ are the Pauli matrices.

For angles ϑ between vectors $\mathbf{B}_0$ and $\mathbf{n}$ not too close to $\pi/2$, the closed electron orbits in momentum space are almost the same for different values of the momentum projection on the magnetic field direction, and the area $S(\varepsilon,p_B)$ 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 $\mathbf{v}=\partial\varepsilon(\mathbf{p})/\partial\mathbf{p}$ of the conduction electrons in the plane of the layers depends weakly on p_B, with an order of smallness $\eta\tan\vartheta$. This means that the energy of the quasiparticles in the one-electron approximation and the Landau correlation function can be expanded in an asymptotic series, the leading term of which is independent of p_B. In the zeroth approximation in the small parameter η the functions $N(\mathbf{p},\mathbf{p}')$ and $S(\mathbf{p},\mathbf{p}')$ can be represented as Fourier series:

$$N(\mathbf{p},\mathbf{p}')=\sum_{n=-\infty}^{\infty} N_n(\varepsilon_F)e^{in(\varphi-\varphi')},$$

$$S(\mathbf{p},\mathbf{p}')=\sum_{n=-\infty}^{\infty} S_n(\varepsilon_F)e^{in(\varphi-\varphi')} \tag{3}$$

with coefficients coupled by the relations $N_{-n}=N_n$, $S_{-n}=S_n$. As variables in $\mathbf{p}$ space we chose the integrals of motion ε and p_B of the charge carriers in the magnetic field and also the phase of the electron velocity, $\varphi=\omega_B t_1$, where t_1 is the time of motion along the trajectory $\varepsilon=\varepsilon_F$, $p_B=\text{const}$. Taking the next terms of the expansion of the correlation function in powers of η into account leads only to negligibly small corrections to the spectrum of the collective modes.

The paramagnetic spin modes are space–time perturbations of the spin density $\mathbf{g}(\mathbf{r},\mathbf{p},t)=\mathrm{Tr}_\sigma(\hat{\sigma}\rho)$. For small deviations from the equilibrium state the spin density can be written as the sum of the equilibrium part $\mathbf{g}_0=-\mu\mathbf{B}_0(\partial f_0/\partial\varepsilon)$ and a small nonequilibrium admixture $-(\partial f_0/\partial\varepsilon)\boldsymbol{\xi}(\mathbf{r},\mathbf{p},t)$, where $f_0(\varepsilon)$ is the Fermi function, $\mu=\mu_0/(1+S_0^{\sim})$, μ_0 is the magnetic moment of a conduction electron, $S_0^{\sim}=\nu(\varepsilon_F)S_0$, and $\nu(\varepsilon_F)$ is the density of states at the Fermi level. The integral of $\mu_0\mathbf{g}_0(\varepsilon)$ over the unit cell in $\mathbf{p}$ space gives the magnetization $\mathbf{M}_0=\chi_0\mathbf{B}_0$ in a uniform static magnetic field with induction $\mathbf{B}_0$, and $\chi_0=\mu_0\mu\nu(\varepsilon_F)$ is the static paramagnetic susceptibility.

According to Ref. 2, the linearized kinetic equation in the case when the perturbation of the spin density $\boldsymbol{\xi}$ is perpendicular to $\mathbf{B}_0$ has the form

$$\frac{\partial\boldsymbol{\xi}}{\partial t}+\left(\mathbf{v}\frac{\partial}{\partial\mathbf{r}}+\frac{e}{c}(\mathbf{v}\times\mathbf{B}_0)\frac{\partial}{\partial\mathbf{p}}\right)\mathbf{\Phi}-\frac{2\mu}{\hbar}[\mathbf{B}_0\times\mathbf{\Phi}]$$
$$-\mu_0\mathbf{v}\frac{\partial\mathbf{B}^{\sim}}{\partial\mathbf{r}}+\frac{2\mu\mu_0}{\hbar}[\mathbf{B}_0\times\mathbf{B}^{\sim}]=I_{\text{coll}}. \tag{4}$$

Here $\mathbf{\Phi}=\boldsymbol{\xi}+\langle S\boldsymbol{\xi}\rangle$, the angle bracket denotes averaging over the Fermi surface,

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

$$\frac{\partial f_0}{\partial\varepsilon}=-\delta(\varepsilon-\varepsilon_F),$$

$\mathbf{B}^{\sim}(r,t)$ is the rf field, e is the electron charge, and c is the speed of light. The collision integral I_{coll} determines two relaxation times: τ_1 and τ_2, the momentum and spin-density relaxation times; $\tau_2\gg\tau_1$. For processes corresponding to the frequency region $kc\gg\omega\gg\tau^{-1}=\tau_1^{-1}+\tau_2^{-1}$ [the wave vector $\mathbf{k}=(k_x,0,k_z)$], the asymptotic behavior of the spectrum of collective modes is completely independent of the specific form of the collision integral.

Expanding the functions $\mathbf{\Phi}=\boldsymbol{\xi}+\langle S\boldsymbol{\xi}\rangle$ and $\boldsymbol{\xi}$ in Fourier series in the variable φ and substituting the results into Eq. (4), we find that the circular components of the renormalized spin density $\Phi^{(\pm)}=\Phi_{x1}\pm i\Phi_y\sim\exp(-i\omega t+i\mathbf{k}\cdot\mathbf{r})$ of the conduction electrons satisfy the integral equations[4]

$$\Phi^{(\pm)}=\int_{-\infty}^{\varphi}d\varphi'\exp\left(\frac{i}{\omega_B}\int_{\varphi'}^{\varphi}d\varphi''(\tilde{\omega}\pm\Omega\right.$$
$$\left.-\mathbf{k}\cdot\mathbf{V}(\varphi'',p_B))\right)\left(i\frac{\mu_0}{\omega_B}(\mathbf{k}\cdot\mathbf{v}(\varphi',p_B)\pm\Omega)B_{\pm}^{\sim}\right.$$
$$\left.-i\frac{\omega}{\omega_B}\sum_{p=-\infty}^{\infty}\lambda_p\overline{\Phi}_p^{(\pm)}e^{ip\varphi'}\right), \tag{5}$$

$\Phi_{x_1}=\Phi_x\cos\vartheta-\Phi_z\sin\vartheta$, the x_1 axis is directed perpendicular to the y axis and to the vector $\mathbf{B}_0$, where $\lambda_p=S_p^{\sim}/(1+S_p^{\sim})$, $\tilde{\omega}=\omega+i0$, $\overline{\mathbf{\Phi}}_p=\langle e^{-ip\varphi}\mathbf{\Phi}\rangle/\langle 1\rangle$, $B_{\pm}^{\sim}=B_{x_1}^{\sim}\pm iB_y^{\sim}$, $\Omega=\omega_s/(1+S_0^{\sim})$, and $\omega_s=-2\mu_0B_0/\hbar$ is the spin paramagnetic resonance frequency.

Multiplying Eq. (5) by $\exp(-in\varphi)$ and integrating with respect to the variables $\beta=p_B/p_0\cos\vartheta$ and φ, we obtain an infinite system of linear equations for the coefficients $\overline{\mathbf{\Phi}}_n^{(\pm)}$ of the Fourier series of the function

$$\langle\Phi^{(\pm)}\rangle_\beta\equiv\frac{1}{2\pi}\int_{-\pi}^{\pi}d\beta\,\Phi^{(\pm)}(\varepsilon_F,\beta,\varphi)$$

$$\sum_{p=-\infty}^{\infty}\left(\delta_{np}-\lambda_p\frac{\omega}{\omega_B}\langle f_{np}(\beta)\rangle_\beta\right)\overline{\Phi}_p^{(\pm)}$$
$$=-\mu_0B_{\pm}^{\sim}\left\langle\frac{1}{2\pi i}\,\frac{\int_0^{2\pi}\int_0^{2\pi}d\varphi d\varphi_1[\mathbf{kv}(\beta,\varphi-\varphi_1)\mp\Omega]\exp[i(p-n)\varphi-ip\varphi_1+iR(\varphi,\varphi_1)]}{1-\exp[2\pi iR(2\pi,2\pi)]}\right\rangle_\beta, \tag{6}$$

$$f_{np}(\beta)=\frac{1}{2\pi i}\,\frac{\int_0^{2\pi}\int_0^{2\pi}d\varphi d\varphi_1\exp[i(p-n)\varphi-ip\varphi_1+iR(\varphi,\varphi_1)]}{1-\exp[2\pi iR(2\pi,2\pi)]}. \tag{7}$$

Here $R(\varphi,\varphi_1)\equiv 1/\omega_B\int_{\varphi-\varphi_1}^{\varphi}d\varphi'[\tilde{\omega}\mp\Omega-\mathbf{k}\cdot\mathbf{v}(\beta,\varphi')]$, and δ_{np} is the Kronecker delta. The dependence of the cyclotron frequency on p_B should be taken into account only in the expression $k_x v_x/\omega_B$ in the argument of the exponential function, under the condition that $\eta k_x v_F\sim\omega_B$.

The coefficients of the Fourier series of the smooth function $\nu(\varepsilon_F)S(\mathbf{p},\mathbf{p}')$ fall off rapidly with increasing summation index, and it is therefore sufficient to keep a finite number of terms of the series in Eqs. (5) and (6). The system of equations (6), together with Maxwell's equation, relates the variable magnetic field and the magnetization and describes the natural oscillations of the spin density in layered conductors with an arbitrary energy spectrum and correlation function. It is easy to see that for finding the spin-wave spectrum it is sufficient to use the homogeneous version of the system of equations (6). We shall neglect in (6) the small inhomogeneous term proportional to $\mu_0 B_\pm^\sim$, which takes into account the influence of the self-consistent field $B_\pm^\sim$. The dispersion relation for the "free" oscillations of the spin density has the form

$$D(\omega^{(0)},\mathbf{k})\equiv\det\left[\delta_{np}-\lambda_p\frac{\omega^{(0)}}{\omega_B}\langle f_{np}(\beta)\rangle_\beta\right]=0. \tag{8}$$

Up to terms proportional to $\chi_0\sim\mu_0^2\nu(\varepsilon_F)$ the frequency ω of the natural oscillations of the magnetization is equal to the frequency $\omega^{(0)}$ of the "free" oscillations of the spin density. At that frequency the magnetic susceptibility has a sharp maximum, and the determinant $D(\omega,\mathbf{k})$ is equal in order of magnitude to χ_0.

The condition that there be no collisionless damping of spin waves reduces to satisfaction of the inequality

$$|\omega-n\omega_B\mp\Omega|>\max|\langle\mathbf{k}\cdot\mathbf{v}\rangle_\varphi|. \tag{9}$$

Outside the region of ω, $\mathbf{k}$ values corresponding to condition (9) the functions $f_{np}(\beta)$ have a pole, and after integration over p_B the dispersion relation acquires a imaginary part responsible for strong absorption of the wave. In layered conductors the electron drift velocity along the magnetic field, $\mathbf{v}_B=\langle\mathbf{v}\rangle_\varphi$, oscillates as a function of the angle ϑ between the magnetic field and the normal to the layers. For certain directions of $\mathbf{B}_0$ with respect to the layers of the conductor $\mathbf{v}_B$ is close to zero, and the damping of the wave is governed by collision processes. Here the existence of collective modes is possible even under the condition $\eta k v_F \gtrsim\omega_B$. In the region of ω and $\mathbf{k}$ values such that $\mathbf{k}\cdot\mathbf{v}_m\gg\omega_B$, $\omega\mp\Omega\ll\mathbf{k}\cdot\mathbf{v}_m$, where $\mathbf{v}_m$ is the maximum value of the velocity in the $\mathbf{k}$ direction, there exist solutions of the dispersion relation (8) in the neighborhood of the resonance

$$\omega=n\omega_B\pm\Omega+\Delta\omega,\Delta\omega\ll\omega_B,n=0,1,2\ldots. \tag{10}$$

Keeping only the first two terms in formula (1) and neglecting anisotropy in the plane of the layers, we write the energy of a quasiparticle in the one-electron approximation as

$$\varepsilon(\mathbf{p})=\frac{p_x^2+p_y^2}{2m}-\eta v_F p_0\cos\frac{p_z}{p_0}, \tag{11}$$

where $v_F=\sqrt{2\varepsilon_F/m}$. The asymptotic solutions accurate to terms of order η for the system of equations of motion corresponding to the dispersion relation (11) are easily found using the standard methods of nonlinear mechanics[5]

$$\begin{aligned}
v_x(t_1)&=v_x^{(0)}(t_1)+v_x^{(1)}(t_1),\quad v_x^{(0)}(t_1)=v_\perp\cos\omega_B(\beta)t_1,\\
v_x^{(1)}(t_1)&=\eta v_F\tan\vartheta J_0(\alpha)\sin\beta\\
&\quad-\eta v_F\tan\vartheta\sum_{n=2}^{\infty}\frac{J_n(\alpha)\sin(\beta-n\pi/2)}{n^2-1}\\
&\quad\times\cos n\omega_B(\beta)t_1,\\
v_z(t_1)&=\eta v_F\sin(\beta-\alpha\cos\omega_B(\beta)t_1).
\end{aligned} \tag{12}$$

Here $\omega_B(\beta)=\omega_B[1+(\eta\tan\vartheta J_1(\alpha)\cos\beta)/2]$ is the cyclotron frequency of quasiparticles with energy (11) in a field

$$\mathbf{B}_0=(B_0\sin\vartheta,0,B_0\cos\vartheta),\quad \omega_B=(|e|B_0/mc)\cos\vartheta,$$

$$\alpha=(mv_F/p_0)\tan\vartheta,$$

$J_n(\alpha)$ is the Bessel function,

$$v_\perp=v_F\left(1-\frac{v_x^{(1)}(0)}{v_F}+\frac{\eta p_0}{mv_F}\cos(\beta-\alpha)\right)$$

is the amplitude of the first harmonic of $v_x(t)$, and the initial phase is chosen such that $v_y(0)=0$.

It follows from relations (12) that

$$\langle\mathbf{kv}\rangle_\varphi=\mathbf{kv}_B=\eta v_F J_0(\alpha)(k_x\tan\vartheta+k_z)\sin\beta. \tag{13}$$

For those directions of $\mathbf{B}_0$ for which α is equal to one of the zeros $\alpha_i=(mv_F/p_0)\tan\vartheta_i$ of the Bessel function $J_0(\alpha)$ the average $\langle\mathbf{k}\cdot\mathbf{v}\rangle_\varphi\sim\eta^2$, and the asymptotic expression for the coefficients $f_{np}(\beta)$ takes the form

$$f_{np}(\beta)=\frac{1}{k_x r_0}\left(\cot\frac{\pi(\omega\mp\Omega)}{\omega_B}\cos\frac{\pi}{2}(n-p)+\frac{\sin\left(R_1(\vartheta_1)+\frac{\pi}{2}(n+p)\right)}{\sin\frac{\pi(\omega\mp\Omega)}{\omega_B}}\right), \tag{14}$$

where

$$\begin{aligned}
R_1(\vartheta_i)&=\int_{-\pi/2}^{\pi/2}\frac{\mathbf{k}\cdot\mathbf{v}(\varphi)}{\omega_B(\beta_i)}d\varphi=2\frac{k_x v_\perp}{\omega_B(\beta_i)}\\
&\quad-\pi\eta\frac{k_z v_F}{\omega_B}H_0(\alpha_i)\cos\beta_i\\
&\quad+\eta\frac{k_x v_F}{2\omega_B}\tan\vartheta_i\cos\beta_i\sum_{n=1}^{\infty}\frac{J_{2n+1}(\alpha_i)}{n(n+1)(2n+1)},
\end{aligned}$$

is the Struve function, $r_0=v_F/\omega_B$, and $\beta_i=p_B/p_0\cos\vartheta_i$. In the case when the correlation function is determined by the zeroth and first Fourier harmonics

$$S(\mathbf{p},\mathbf{p}')=S_0+2S_1\cos(\varphi-\varphi'),$$

the solution of dispersion relation (8) is determined by formula (10) with

$$\Delta\omega = \frac{n\omega_B \pm \Omega}{\pi k_x r_0}\gamma_{1,2}.$$

It is easy to obtain from (8) a quadratic equation for $\gamma_{1,2}$, the roots of which are

$$\gamma_{1,2} = \frac{1}{2}[\lambda_0 + 2\lambda_1 + (-1)^n(\lambda_0 - 2\lambda_1)g \pm ((\lambda_0 + 2\lambda_1 + (-1)^n(\lambda_0 - 2\lambda_1)g)^2 + 8\lambda_0\lambda_1(-1 + g^2 + h^2))^{1/2}],$$

where $g = \langle \sin R_1(\vartheta_i)\rangle_\beta$, and $h = \langle \cos R_1(\vartheta_i)\rangle_\beta$.

In the short-wavelength limit for the selected directions of the external magnetic field there exist spin waves with frequencies (10) close to the resonance frequencies $\omega_r = n\omega_B \pm \Omega$. The correction (15) to the resonance frequency is a rapidly oscillating function of wave number. An analogous type of excitations exists in quasi-isotropic metals only when the direction of wave propagation is strictly perpendicular to $\mathbf{B}_0$.

*E-mail: stepanenko@ilt.kharkov.ua

---

[1] J. Singelton, Rep. Prog. Phys. **63**, 1111 (2000).
[2] V. P. Silin, Zh. Eksp. Teor. Fiz. **35**, 1243 (1958) [Sov. Phys. JETP **8**, 870 (1959)].
[3] S. Schultz and G. Dunifer, Phys. Rev. Lett. **18**, 283 (1967).
[4] V. G. Peschansky and D. I. Stepanenko, JETP Lett. **78**, 322 (2003).
[5] N. N. Bogolyubov and Yu. A. Mitropol'skiĭ, *Asymptotic Methods in the Theory of Nonlinear Oscillations*, translation of 2nd Russ. ed., Gordon and Breach, New York (1962), cited 4th Russ. ed., Nauka, Moscow (1974).

Translated by Steve Torstveit