

High-frequency resonances and weakly damped collective modes in highly anisotropic Q1D conductors

Yu. A. Kolesnichenko V. G. Peschansky D. I. Stepanenko

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High-frequency resonances and weakly damped collective modes in highly anisotropic Q1D conductors

Yu. A. Kolesnichenko

B. I. Verkin Institute of Low-temperature Physics and Engineering, National Academy of Sciences of Ukraine, pr. Nauki 47, Kharkov 61103, Ukraine

V. G. Peschansky

B. I. Verkin Institute of Low-temperature Physics and Engineering, National Academy of Sciences of Ukraine, pr. Nauki 47, Kharkov 61103, Ukraine and V. N. Karazin Kharkov National University, pl. Svobody 4, Kharkov 61077, Ukraine

D. I. Stepanenko^{a)}

B. I. Verkin Institute of Low-temperature Physics and Engineering, National Academy of Sciences of Ukraine, pr. Nauki 47, Kharkov 61103, Ukraine

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It is shown that weakly damped electromagnetic waves with polarization perpendicular to the direction of highest conductivity can propagate in highly anisotropic organic conductors of the quasi-one dimensional type in a magnetic field. The dispersion relations are analyzed numerically and simple analytic expressions are obtained for the spectrum of the collective modes in a number of limiting cases. *Published by AIP Publishing.* [<http://dx.doi.org/10.1063/1.4977209>]

1. Introduction

A variety of electromagnetic collective modes, some of which are analogous to those in gaseous plasmas, can exist in metals at low temperatures.^{1,2} Most of these excitations are strongly damped, and weakly damped waves can only exist within a few frequency regions and for certain parameters of the solid state plasma. Without a magnetic field, electromagnetic waves with frequency ω lower than the plasma frequency ω_p cannot propagate in plasma media; they are damped or fully reflected. Wave absorption is caused by electron collisions and collisionless Landau damping, which is a resonant absorption of an electromagnetic field by charge carriers with velocities along the wave vector equal to the phase velocity of the wave. The latter mechanism for absorption is the basic mechanism for low frequency modes with $\omega\tau \gg 1$ (where τ is the mean free time for the electrons). When $\omega > \omega_p$ the displacement current exceeds the conduction current, the dielectric constant is positive in the collisionless limit $\tau \rightarrow \infty$, and the plasma is transparent for electromagnetic waves.

A magnetic field affects the electron dynamics and changes the electromagnetic properties of a plasma medium. At low temperatures, it is possible for waves to propagate in conductors in a magnetic field at frequencies substantially below ω_p with a damping mean free path equal to the mean free path of the charge carriers under conditions such that an electron completes at least a few cyclotron orbits during its mean free time. There is yet another mechanism for collision absorption when a constant magnetic field is present: cyclotron damping, which occurs when the frequency of the electromagnetic field is equal to the cyclotron frequency of the conduction electrons. Electrons moving along spirals in phase with the wave are accelerated in the electric field in a plane perpendicular to \mathbf{H}_0 and absorb energy from the electromagnetic field.

As a rule, weakly damped waves are associated with high-frequency resonances. The electromagnetic energy absorbed under resonance conditions can propagate in the form of collective modes. The high-frequency resonances appear because of the periodic motion of conduction electrons in a magnetic field along the Fermi surface (FS) when their mean free time is long enough. In highly anisotropic organic conductors, resonances can arise from the dynamics of quasi-two dimensional (Q2D), as well as quasi-one dimensional (Q1D), groups of charge carriers.^{3–16} We have previously examined weakly damped intrinsic modes in Q2D and Q1D low-dimensional conducting systems under conditions of strong spatial dispersion, when the conductivity can be calculated analytically by the stationary phase method.^{17–19} Weakly damped electromagnetic waves polarized in the direction of highest conductivity have been studied²⁰ in highly anisotropic organic conductors with Fermi surfaces in the form of two weakly corrugated planes. This paper discusses electromagnetic waves polarized perpendicular to the conducting chain in Q1D-type organic conductors, which can appear when there is a nonlocal coupling between the electric current and the variable electromagnetic field. A numerical analysis of the dispersion relations yields a fairly complete idea of the dispersion of weakly damped electromagnetic waves. Analytic expressions are obtained for the spectrum of the weakly damped modes in a number of limiting cases.

2. Resonances in the high-frequency conductivity

The main structural components of Q1D conductors are organic molecules or molecular complexes, such as tetrathyltetraselenafulvalene (TMTSF), tetracyanoquinodimethene (TCNQ), di-methylselylenedithiodiselenadithiafulvalene (DMET), etc., with donor or acceptor properties. The non-

radicals of these molecules form regular stacks along a defined direction. The electrical conductivity along the stacks is several orders of magnitude higher than that in the transverse direction. The best known examples of conductors of the Q1D type with a highly anisotropic Fermi surface are the so-called Bechgard salts $(\text{TMTSF})_2\text{X}$ (X denotes a set of different anions). Although all these substances have a complex chemical structure, they have a fairly simple Fermi surface that can be sketched as a pair of weakly corrugated surfaces, as in Fig. 1.

Usually the electron energy spectrum corresponding to this kind of FS can be written in the form

$$\varepsilon(\mathbf{p}) = v_F(|p_x| - p_F) + B \cos \frac{p_y}{p_2} + C \cos \frac{p_z}{p_3}, \quad (1)$$

where $v_F = (A/p_1) \sin(p_F/p_1)$ and p_F are the velocity and momentum on the FS in the direction of the maximum conductivity; A , B , and C are overlap integrals that obey $A \gg B \gg C$; the constants $p_1 = \hbar/a_1$, $p_2 = \hbar/a_2$, and $p_3 = \hbar/a_3$ are determined by the principal lattice periods a_1, a_2, a_3 ; and \hbar is the Planck constant. The characteristic values of the overlap integrals are usually of order $A \sim 0.5 \text{ eV}$, $B \sim 0.05 \text{ eV}$, and $C \sim 2 \text{ meV}$. The dispersion relations (1) correspond to the energy spectrum in the strong coupling approximation, linearized in the direction of maximum conductivity along the Fermi level ε_F .

Without quantization of the electron energy levels in a magnetic field, for frequencies ω of the electromagnetic field below C/\hbar the kinetic properties of the conductor can be described in a quasiclassical approximation. In the case when the magnetic field $\mathbf{H}_0 = (0, H_0 \sin \vartheta, H_0 \cos \vartheta)$ is perpendicular to the direction of the conducting chain, the components of the electron velocity are given by

$$\begin{aligned} v_x &= \text{sign}(p_x)v_F, & v_y &= \text{sign}(p_x)v_2 \sin \Omega t \\ v_z &= v_3 \sin \left(\frac{p_H}{p_3 \cos \vartheta} - \text{sign}(p_x)\alpha\Omega t \right), \end{aligned} \quad (2)$$

where $\Omega = (|e|v_F H_0 / c p_2) \cos \vartheta \equiv \Omega_0 \cos \vartheta$ is the analog of the cyclotron frequency for electrons with the dispersion law (1); $v_2 = B/p_2$ and $v_3 = C/p_3$ are the characteristic velocities of the electrons in a plane perpendicular to the conducting chain; $\alpha = (p_2/p_3) \text{tg} \vartheta$ and $p_H = (\mathbf{p}\mathbf{H}_0)/H_0$ are the projection of the momentum on the direction of the magnetic

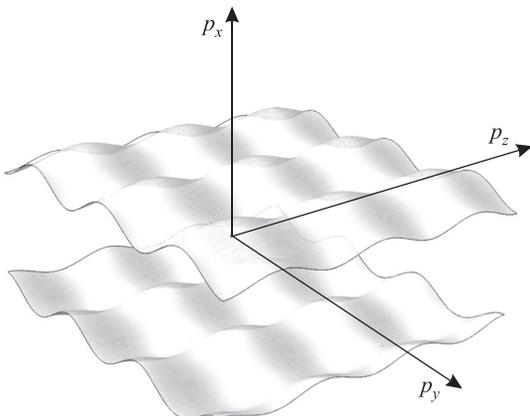


FIG. 1. A Q1D Fermi surface in the form of a pair of weakly corrugated planes: the x axis is the direction of highest conductivity.

field; and e is the electronic charge. The value of $(p_x) = \pm 1$ corresponds to the different sheets of the FS.

The electrical conductivity tensor can be written as follows:

$$\begin{aligned} \sigma_{ij}(\omega, \mathbf{k}) &= \frac{2|e|^3 H_0}{(2\pi\hbar)^3 c} \sum_{\text{sign}(p_x)=\pm 1} \int dp_H \int_0^{2\pi/\Omega} dt v_i(t) \\ &\times \int_{-\infty}^t dt' v_j(t') \exp \left(i\tilde{\omega}(t-t') - i \int_t^{t'} dt'' \mathbf{k}\mathbf{v}(t'') \right), \end{aligned} \quad (3)$$

where $\tilde{\omega} = \omega + i\tau^{-1}$. As the variables in momentum space we have chosen the integrals of motion ε , p_H , and t —the time the electron moves in the magnetic field. The sign of the sums with respect to $\text{sign}(p_x) = \pm 1$ in Eq. (3) signifies summing over the sheets of the FS.

Let us consider the case when the wave vector $\mathbf{k} = (0, k \sin \phi, k \cos \phi)$ is orthogonal to the direction of the maximum conductivity. For the energy spectrum (1) and this geometry for the problem, the components of the tensor σ_{ij} take the form

$$\sigma_{xx} = \frac{\omega_0^2}{2\pi\Omega} \int_0^\infty d\varphi e^{i\frac{\omega_0}{\Omega}\varphi} J_0 \left(2Y \sin \frac{\alpha\varphi}{2} \right) J_0 \left(2X \sin \frac{\varphi}{2} \right), \quad (4)$$

$$\begin{aligned} \sigma_{yy} &= \frac{\omega_0^2}{4\pi\Omega} \left(\frac{v_2}{v_F} \right)^2 \int_0^\infty d\varphi e^{i\frac{\omega_0}{\Omega}\varphi} J_0 \left(2Y \sin \frac{\alpha\varphi}{2} \right) \\ &\times \left(J_0 \left(2X \sin \frac{\varphi}{2} \right) \cos \varphi - J_2 \left(2X \sin \frac{\varphi}{2} \right) \right), \end{aligned} \quad (5)$$

$$\begin{aligned} \sigma_{zz} &= \frac{\omega_0^2}{4\pi\Omega} \left(\frac{v_3}{v_F} \right)^2 \int_0^\infty d\varphi e^{i\frac{\omega_0}{\Omega}\varphi} J_0 \left(2X \sin \frac{\varphi}{2} \right) \\ &\times \left(J_0 \left(2Y \sin \frac{\alpha\varphi}{2} \right) \cos \alpha\varphi - J_2 \left(2Y \sin \frac{\alpha\varphi}{2} \right) \right), \end{aligned} \quad (6)$$

$$\begin{aligned} \sigma_{yz} = \sigma_{zy} &= -\frac{\omega_0^2}{2\pi\Omega} \frac{v_2 v_3}{v_F} \times \int_0^\infty d\varphi e^{i\frac{\omega_0}{\Omega}\varphi} J_1 \left(2Y \sin \frac{\alpha\varphi}{2} \right) \\ &\times J_1 \left(2X \sin \frac{\varphi}{2} \right) \cos \frac{\varphi}{2} \cos \frac{\alpha\varphi}{2}, \\ \sigma_{xy} = \sigma_{yx} &= \sigma_{xz} = \sigma_{zx} = 0, \end{aligned} \quad (7)$$

where $X = k_y v_2 / \Omega$, $Y = k_z v_3 / (\alpha\Omega)$, and $J_n(x)$ is the Bessel function of n -th order. For values of the overlap integral $A \sim 0.5 \text{ eV}$, the frequency $\omega_0 = (4e^2 p_2 p_3 v_F / \hbar^3)$ is on the order of 10^{15} s^{-1} .

It is easy to see that the oscillations in the electron velocity components v_y and v_z lead yield the resonances in the high-frequency conductivity. Expanding the Bessel functions in Eqs. (4)–(6) in Fourier series in φ and $\alpha\varphi$, i.e., taking

$$\begin{aligned} J_0(2Z \sin(\psi/2)) &= \sum_{n=-\infty}^{\infty} J_n^2(Z) \exp(in\psi), \\ J_0(2Z \sin(\psi/2)) &= \sum_{n=-\infty}^{\infty} J_{1-n}(Z) J_{1+n}(Z) \exp(in\psi), \\ Z &= \{X, Y\}, \quad \psi = \{\varphi, \alpha\varphi\}, \end{aligned}$$

and integrating with respect to φ , for the diagonal components of the conductivity we obtain the following:

$$\sigma_{xx} = \frac{i\omega_0^2}{2\pi} \sum_{n,m=-\infty}^{\infty} \frac{J_n^2(X)J_m^2(Y)}{\tilde{\omega} - n\Omega - \alpha m\Omega}, \quad (8)$$

$$\sigma_{yy} = \frac{i\omega_0^2}{4\pi} \left(\frac{v_2}{v_F}\right)^2 \times \sum_{n,m=-\infty}^{\infty} \frac{J_m^2(Y)(J_{n-1}^2(X) + J_{n+1}^2(X) + 2J_{n-1}(X)J_{n+1}(X))}{\tilde{\omega} - n\Omega - \alpha m\Omega}, \quad (9)$$

$$\sigma_{zz} = \frac{i\omega_0^2}{4\pi} \left(\frac{v_3}{v_F}\right)^2 \times \sum_{n,m=-\infty}^{\infty} \frac{J_n^2(X)(J_{m-1}^2(Y) + J_{m+1}^2(Y) + 2J_{m-1}(Y)J_{m+1}(Y))}{\tilde{\omega} - n\Omega - \alpha m\Omega}. \quad (10)$$

When the mean free time τ is long enough, i.e., $\Omega_0\tau \gg 1$, local maxima in the high-frequency conductivity and the microwave absorption appear under the condition

$$\omega - n\Omega - \alpha m\Omega = 0. \quad (11)$$

Because of the motion of the electrons in the z direction, however, the resonances at frequencies $\omega = m\alpha\Omega \equiv m\Omega$ can appear only for a short wavelength of the electromagnetic field, such that $Y^2\alpha\Omega\tau$ is comparable to unity.

In the collisionless limit $\tau \rightarrow \infty$, the high-frequency conductivity may be nondissipative. As a result, weakly damped collective modes may appear in Q1D-type conductors with a strong anisotropy in the FS.

3. Collective mode spectra

Assuming that the time dependence for all the variables is in the form $(i\mathbf{k}\mathbf{r} - i\omega t)$, the Maxwell equations easily yield a dispersion relation that gives the frequency $\omega(\mathbf{k})$ of the eigenmodes of the electromagnetic field

$$D \equiv \det \left[k^2 \delta_{ij} - k_i k_j - \frac{\omega^2}{c^2} \varepsilon_{ij}(\omega, \mathbf{k}) \right] = 0. \quad (12)$$

The real parts of the roots of Eq. (12) determine the spectrum of the collective modes and the imaginary parts, the damping decrement. Here $\varepsilon_{ij}(\omega, \mathbf{k}) = \delta_{ij} + (4\pi/\omega)\sigma_{ij}(\omega, \mathbf{k})$ is the dielectric conductivity tensor and δ_{ij} is the Kronecker symbol. For frequencies ω much lower than σ_{ij} , the first term in the expression for ε_{ij} can be neglected.

We now consider the case where the wave vector $\mathbf{k} = (0, k, 0)$ is directed along the y axis. The dispersion relation (12) factorizes into

$$D = \left(k^2 - \frac{\omega}{c^2} \varepsilon_{xx}(\omega, \mathbf{k}) \right) \left(-\frac{\omega^2}{c^2} \varepsilon_{yy}(\omega, \mathbf{k}) \right) \times \left(k^2 - \frac{\omega^2}{c^2} \varepsilon_{zz}(\omega, \mathbf{k}) \right) = 0, \quad (13)$$

and breaks up into three equations. The first, which describes the transverse mode with the electric field polarized in the direction of the maximum conductivity, has been studied in detail elsewhere²⁰ for arbitrary orientations of the magnetic field and the wave vector. The second has a weakly damped solution in the frequency range $\omega > (B/\varepsilon_F)^2\omega_0$ and determines the longitudinal plasma oscillations in the y direction. The third equation

$$k^2 - \frac{\omega^2}{c^2} \varepsilon_{zz}(\omega, \mathbf{k}) = 0, \quad (14)$$

describes the collective mode with an electric field polarized in the direction of minimal conductivity. When $k_z = 0$, the integral expression (6) for the components of the conductivity can be simplified to

$$\sigma_{zz} = \frac{i\omega_0^2}{4\alpha\Omega} \left(\frac{v_3}{v_F}\right)^2 \left[\frac{J_{(\tilde{\omega}-\alpha\Omega)/\Omega}(X)J_{-(\tilde{\omega}-\alpha\Omega)/\Omega}(X)}{\sin\left(\pi\frac{\tilde{\omega}-\alpha\Omega}{\Omega}\right)} + \frac{J_{(\tilde{\omega}+\alpha\Omega)/\Omega}(X)J_{-(\tilde{\omega}+\alpha\Omega)/\Omega}(X)}{\sin\left(\pi\frac{\tilde{\omega}+\alpha\Omega}{\Omega}\right)} \right]. \quad (15)$$

The spectrum of the collective modes can be written in analytic form in the short- and long-wavelength limits. In the case of a weak spatial dispersion $X \ll 1$, Eq. (15) can be expanded in a rapidly decreasing series and Eq. (14) becomes algebraic. In the main approximation with respect to X^2 , the dispersion relation for the low-frequency mode is given by

$$\omega = \frac{\alpha\Omega}{\sqrt{2}} \frac{kc v_F}{\omega_0 v_3}. \quad (16)$$

For relatively large values $X \gg 1$, Eq. (14) can be simplified using an asymptotic representation of the Bessel function in Eq. (15) as a trigonometric function. For small values of the parameter $(1/\pi X^3)(\omega_0 v_2 v_3)^2 / (v_F \Omega c)^2 \ll 1$, the eigenfrequencies are close to the resonance frequencies $\omega = (n \pm \alpha)\Omega$, i.e.

$$\omega = (n \pm \alpha)\Omega \left(1 - \frac{1}{\pi X^3} \left(\frac{\omega_0 v_2}{\Omega c}\right)^2 \left(\frac{v_3}{v_F}\right)^2 (1 - (-1)^n \sin 2X) \right). \quad (17)$$

The deviation of ω from the resonance frequency $(n \pm \alpha)\Omega$ oscillates as $\sin 2X$ and falls off with k as k^{-3} .

For arbitrary values of the dimensionless component of the wave vector X , solutions of the transcendental Eq. (14) cannot be obtained in analytic form. Numerical calculations of the spectra of the collective modes in the limit of large relaxation times are shown in Fig. 2. This figure shows that the weakly damped waves vanish when the wave frequency is close to the resonance frequency $\omega = (n \pm \alpha)\Omega$ because of strong cyclotron absorption.

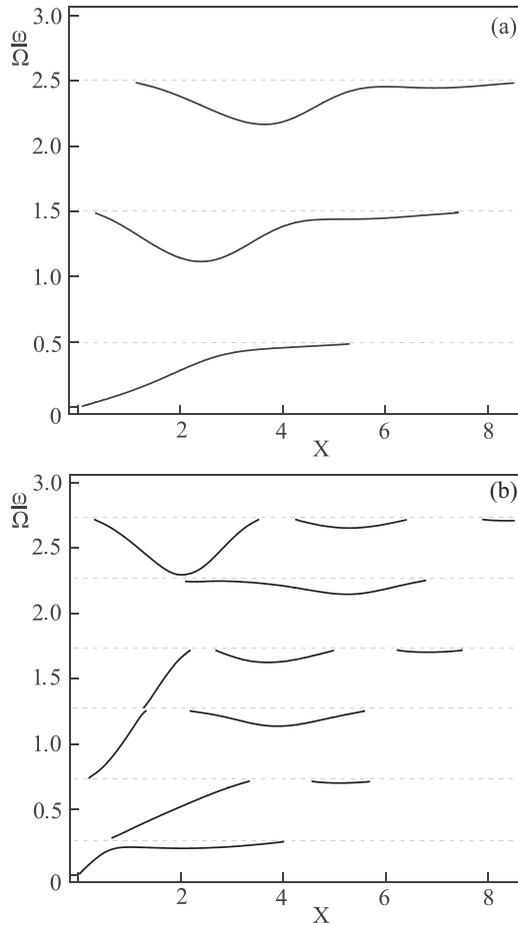


FIG. 2. Collective mode spectra for $\mathbf{k} = (0, k, 0)$, $(\frac{\omega_0 v_2 v_3}{\alpha \Omega v_F c})^2 = 10$, $(\Omega \tau)^{-1} = 0, 01$; $\alpha = 1/2$ (a), $\alpha = \sqrt{3}$ (b). The dotted lines correspond to the resonance frequencies $\omega = (n \pm \alpha)\Omega$.

When the wave vector $\mathbf{k} = (0, 0, k)$ is parallel to the z axis, the dispersion relation transforms to

$$D = \left(k^2 - \frac{\omega^2}{c^2} \varepsilon_{xx}(\omega, \mathbf{k}) \right) \left(k^2 - \frac{\omega^2}{c^2} \varepsilon_{yy}(\omega, \mathbf{k}) \right) \times \left(\frac{\omega^2}{c^2} \varepsilon_{zz}(\omega, \mathbf{k}) \right) = 0. \quad (18)$$

The equation

$$k^2 - \frac{\omega^2}{c^2} \varepsilon_{yy}(\omega, \mathbf{k}) = 0, \quad (19)$$

describes a collective mode with an electric field polarized in the y direction. After calculating the integral in Eq. (5), for the components σ_{yy} of the conductivity we obtain

$$\sigma_{yy} = \frac{i\omega_0^2}{4\alpha\Omega} \left(\frac{v_2}{v_F} \right)^2 \left[\frac{J_{(\tilde{\omega}-\Omega)/\alpha\Omega}(Y) J_{-(\tilde{\omega}-\Omega)/\alpha\Omega}(Y)}{\sin\left(\pi \frac{\tilde{\omega}-\Omega}{\alpha\Omega}\right)} + \frac{J_{(\tilde{\omega}+\Omega)/\alpha\Omega}(Y) J_{-(\tilde{\omega}+\Omega)/\alpha\Omega}(Y)}{\sin\left(\pi \frac{\tilde{\omega}+\Omega}{\alpha\Omega}\right)} \right]. \quad (20)$$

For $Y \ll 1$, Eq. (19) can be simplified using an asymptotic expansion of Eq. (20) as a power series in Y . As a result, we find the dispersion relation for the low-frequency mode

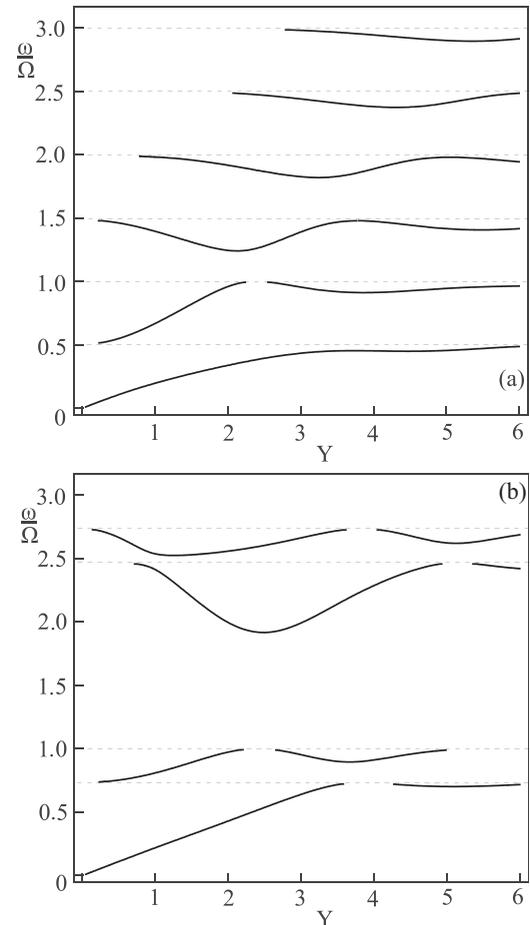


FIG. 3. Collective mode spectra for $\mathbf{k} = (0, 0, k)$, $(\frac{\omega_0 v_2 v_3}{\alpha \Omega v_F c})^2 = 10$, $(\Omega \tau)^{-1} = 0, 01$; $\alpha = 1/2$ (a), $\alpha = \sqrt{3}$ (b). The dotted lines correspond to the resonance frequencies $\omega = (n \pm \alpha)\Omega$.

$$\omega = \frac{\Omega}{\sqrt{2}} \frac{kc v_F}{\omega_0 v_2}. \quad (21)$$

Spectra of waves propagating in the direction of minimal conductivity are shown in Fig. 3.

The necessary condition for the appearance of weakly damped resonance modes, as for other high-frequency resonance effects in a magnetic field, is $\Omega_0 \tau \gg 1$. The numerical calculations shown above actually correspond to the collisionless limit $\tau^{-1} \rightarrow 0$. The effect of electron collisions on the wave process leads to strong damping of the wave neighborhoods of order τ^{-1} near the resonance frequencies Ω_r . For ω such that $|\omega - n\Omega_r| < \tau^{-1}$, the diagonal components of the conductivity have a larger real part that is responsible for strong absorption of the wave. In this region, there are no weakly damped waves.

Collective waves with frequencies near harmonics of the resonance frequency can occur when there is a nonlocal coupling between the current and the variable electric field. The dispersion effects are more substantial at high frequencies.

4. Conclusion

The weakly damped eigenmodes are collective excitations of Bose type in the electron plasma of solids. Electromagnetic modes in highly anisotropic conductors with a Q1D electron energy spectrum are related to resonant high-frequency conductivity in a strong magnetic field with

almost no collisions through the corrugated sheets of the Fermi surface. The components v_y and v_z of the electron velocity oscillate at frequencies Ω and $\Omega_1 = (p_2/p_3)\text{tg } \vartheta \Omega$, respectively, and generate resonances in the kinetic coefficients of conductors. Thus, resonances in the high-frequency conductivity can occur at two resonance frequencies and their harmonics. Resonances owing to motion of the electrons in the direction of the minimal conductivity can show up only for electromagnetic fields with wavelengths short enough that $(kv_3/\Omega_1)^2$ is comparable to unity. Spatial dispersion is a necessary condition for the existence of electromagnetic waves with frequencies near the resonances, and dispersion effects become more significant with increasing ω . The effect of electron collisions on the wave process leads to vanishing of the weakly damped collective modes in neighborhoods of order τ^{-1} near the resonance frequencies because of strong cyclotron absorption.

^{a)}Email: stepanenko@ilt.kharkov.ua

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