

## Cyclotron resonance in quasi-two-dimensional metals in a tilted magnetic field

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The cyclotron resonance in a tilted magnetic field in quasi-two-dimensional organic metals is studied under conditions of strong spatial dispersion. It is shown that, as opposed to ordinary metals in quasi-two-dimensional conductors, a periodic dependence of the impedance on the reciprocal of the magnetic field shows up in the first approximation with respect to the small parameter equal to the ratio of the depth of the skin layer to the electron Larmor radius. Under resonance conditions in the collisionless limit the conductivity has a square-root singularity, while the amplitude of the oscillations in the impedance increases as the anisotropy parameter of the Fermi surface decreases.

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In 1957, A. A. Galkin and P. A. Bezuglyi discovered the Azbel-Kaner cyclotron resonance<sup>1</sup> in tin and lead.<sup>2,3</sup> Small resonance peaks were observed against the background of a smooth dependence of the absorption of electromagnetic waves on a sufficiently high magnetic field  $\mathbf{H}$  parallel to the sample surface. These papers<sup>1–3</sup> stimulated intense study of the resonance absorption of microwaves in metals in many countries. Experimental studies of resonance high-frequency phenomena turned out to be a reliable spectroscopic technique for determining the characteristics of the electron energy spectrum of metals. According to the Azbel-Kaner theory<sup>1</sup> the resonance field  $H_{\text{res}}$  yields important information on the extreme effective mass  $m^*$  of charge carriers with the Fermi energy  $\varepsilon_F$ . Under the conditions of the anomalous skin effect, when the depth  $\delta$  of the skin layer is considerably smaller than either the mean free path of the conduction electrons or the diameter of their electron orbit  $2r_0$ , a magnetic field parallel to the sample surface will repeatedly return the charge carriers to the skin layer. In this case the conduction electrons interact resonantly with a high frequency electromagnetic field when the frequency  $\omega$  of the field is a multiple of the cyclotron orbital frequency  $\omega_H = |e|H/(m^*c)$  ( $e$  is the electronic charge and  $c$  is the speed of light) of the electrons. In a magnetic field that is tilted by a small angle relative to the sample surface, the resonance frequencies undergo Doppler splitting owing to drift of the charge carriers in the depth of the metal. When the tilt of the field is large, the resonance vanishes in the first approximation with respect to the small parameter  $\delta/r_0$ , i.e., the oscillatory dependence of the impedance on  $H^{-1}$  shows up only in higher order terms with respect to  $\delta/r_0$ .<sup>4</sup>

In low-dimensionality conducting structures, such as organic wires based on tetrathiafulvalene, dichalcogenides of the transition metals, graphite, etc., resonance phenomena during absorption of electromagnetic waves with lengths comparable to the charge carrier mean free path show up more distinctly, since almost all the electrons on the Fermi surface (FS) are involved in their formation and not a

isolated group, as in ordinary metals. Among materials of this type, organic wires occupy an important place, with their complicated chemical composition and a distinct quasi-one- or -two-dimensional anisotropy in their kinetic coefficients owing to their crystal structure. The best known examples of these materials are the salts of tetrathiafulvalene (TTF), bis(ethylenedithio) tetrathiafulvalene (BEDT-TTF), and tetraselenatetracene (TST). Interest in low-dimensional wires of organic origin arises from several of their distinctive properties, such as low dimensionality, a large variety of phase states, the existence of a superconducting phase, and the possibility of varying the ground state by means of comparatively weak external interactions. In a number of organic compounds (e.g., the BEDT-TTF salts) ion radicals of the organic molecules form conducting layers alternating with layers of counter ions. Experimental observations of high frequency resonances<sup>5–10</sup> and the Shubnikov-de Haas and de Haas-van Alphen effects<sup>11–14</sup> in layered organic structures indicates that their electrical conductivity is caused by a group of fermions analogous to the conduction electrons in ordinary metals. The FS of these conductors is open with slight corrugation along the normal to the layers; it can be in the form of multiple sheets and consist of topologically different elements, such as cylinders and planes. Studies of galvanomagnetic effects<sup>15–17</sup> show that the Fermi surfaces of the salts (BEDT-TTF)<sub>2</sub>IBr<sub>2</sub> and (BEDT-TTF)<sub>2</sub>I<sub>3</sub> consist of just a single slightly corrugated cylinder. The anisotropy of the FS is characterized by a small parameter  $\eta$  that is on the order of the ratio of the characteristic velocities of the electrons along the normal to the layers and in the plane of the layers.

High frequency absorption in organic wires in high magnetic fields differs in several ways because of the strong anisotropy of the FS. Collisionless absorption is caused by electrons whose velocity  $\mathbf{v}$  satisfies the equation

$$\omega - n\omega_H - \mathbf{k}\mathbf{v}_D = 0, \quad (1)$$



where  $\mathbf{k}$  is the wave vector,  $\mathbf{v}_D = \langle \mathbf{v} \rangle$  is the drift velocity, the bracket  $\langle \dots \rangle$  denotes averaging over the period  $T = 2\pi/\omega_H$  of the electron motion in the magnetic field. Drift of the electrons leads to a Doppler shift in the resonance frequencies for a varying electromagnetic field and to Landau damping. If  $\mathbf{k}\mathbf{v}_D \ll \omega_H$ , then Landau damping can be neglected and the resonance absorption is independent of the direction of the magnetic field. Although  $\mathbf{v}_D$  is on the order of the characteristic velocity in the direction of the lowest conductivity, the displacement of an electron over the period  $T$  can exceed the skin depth  $\delta \propto k^{-1}$ . In that case  $\mathbf{k}\mathbf{v}_D$  is on the order of  $\omega_H$  and the electron drift has a significant influence on resonance absorption of a high frequency field. In particular, the position of the Landau absorption regions depends on the orientation of  $\mathbf{H}_0$ , since  $\mathbf{v}_D$  is an oscillating function of the angle  $\vartheta$  between the magnetic field and the direction of the minimal conductivity. As a result, the conditions for strong spatial dispersion now include angular oscillations of the high frequency conductivity in the plane of the layers and of the surface impedance owing to the angular dependence of the electron drift velocity.<sup>18</sup> In this paper we study the resonance absorption of high-frequency electromagnetic fields in organic metals with different values of the FS anisotropy parameter  $\eta$  under conditions such that Landau damping is significant. A periodic dependence shows up in the first approximation with respect to the small parameter  $\delta/r_0$ . Under resonance conditions in the collisionless limit, the conductivity has a square root singularity and the amplitude of the oscillations in the impedance increases as  $\eta$  is reduced.

We choose a coordinate system  $XYZ$  such that the  $z$  axis is parallel to the direction of the minimal conductivity and the  $x$  axis is perpendicular to  $\mathbf{H}_0 = (0, H_0 \sin \vartheta, H_0 \cos \vartheta)$  and the wave vector  $\mathbf{k} = (0, k \sin \phi, k \cos \phi)$ . We also use another coordinate system  $x\xi\zeta$  in which the  $\xi$  axis is parallel to  $\mathbf{k}$ .

The electric field inside the wire is determined by the Maxwell equation

$$\frac{d^2 E_i(\xi)}{d\xi^2} = \frac{4\pi i\omega}{c^2} j_i(\xi), \quad (2)$$

and by the condition that current does not flow through the boundary of the sample

$$j_\xi(\xi) = 0,$$

which follows from the equation  $\text{div} \mathbf{j} = 0$ . Equation (2) must be supplemented by a kinetic equation for the distribution function of the conduction electrons and the material equations which relate the current density to the electric field.

We continue the electric field  $\mathbf{E}(\xi, t) = \mathbf{E}(\xi)e^{-i\omega t}$  and current density  $\mathbf{j}(\xi, t) = \mathbf{j}(\xi)e^{-i\omega t}$  evenly into the region  $\xi < 0$  outside the conductor. The equations for the Fourier components are

$$k^2 \mathcal{E}_i(k) + 2E'_i(0) = \frac{4\pi i\omega}{c^2} j_i(k),$$

$$\left\{ \begin{array}{l} \mathcal{E}_i(k) \\ j_i(k) \end{array} \right\} = 2 \int_0^\infty \left\{ \begin{array}{l} E_i(\xi) \\ j_i(\xi) \end{array} \right\} \cos k\xi d\xi, \quad (3)$$

Here we use the fact that the derivative of the electric field at the surface of the conductor undergoes a jump from  $E'_i(0)$  to  $-E'_i(0)$ .

In a tilted magnetic field the resonance part of the current density is produced by electrons that do not collide with the boundary of the sample. Even for purely mirror reflection the projection  $p_H$  of the momentum is not conserved and after colliding with the surface an electron goes into another orbit. If  $\eta r_0 \simeq \delta$ , then there are no electrons which can periodically be reflected from the conductor surface as they move (the so-called "jump" trajectories) and thereby generate a resonance in the high-frequency conductivity. For this reason we neglect the contribution to the conductivity tensor  $\sigma_{ij}$  of the electrons that collide with the surface and write the Fourier component of the current density in the form

$$j_i(k) = \sigma_{ij}(k) \mathcal{E}_j(k), \quad (4)$$

where

$$\begin{aligned} \sigma_{ij}(k) = & \frac{2|e|^3 H_0}{(2\pi\hbar)^3 c} \\ & \times \int dp_H \left[ 1 - \exp \left( i \frac{2\pi}{\omega_H} \tilde{\omega} - i \int_0^{2\pi/\omega_H} dt' \mathbf{k}\mathbf{v}(t') \right) \right]^{-1} \\ & \times \int_0^{2\pi/\omega_H} dt v_i(t) \int_0^{2\pi/\omega_H} dt_1 v_j(t-t_1) \\ & \times \exp \left( i\tilde{\omega}t_1 - i \int_{t-t_1}^t dt' \mathbf{k}\mathbf{v}(t') \right). \end{aligned} \quad (5)$$

Here  $\tilde{\omega} = \omega + i/\tau$  and  $\tau$  is the mean free time of the electrons. Under resonance conditions  $\omega \approx n\omega_H$ ,  $\omega_H\tau \gg 1$ , and in a magnetic field parallel to the conductor surface the current density (4) exceeds the current density owing to electron collisions with the surface by a factor of  $B\omega_H\tau$ ; that is, in this case Eq. (4) will be asymptotically accurate.

On eliminating the Fourier component of the longitudinal current,  $\mathcal{E}_\xi(k)$ , from Eq. (3), we obtain

$$\begin{aligned} D_{ij} \mathcal{E}_j(k) = & \left\{ k^2 \delta_{ij} - \frac{4\pi i\omega}{c^2} \left( \sigma_{ij}(k) - \frac{\sigma_{i\xi}(k) \sigma_{\xi j}(k)}{\sigma_{\xi\xi}(k)} \right) \right\} \\ & \times \mathcal{E}_j(k) = 2E'_i(0), \end{aligned} \quad (6)$$

where  $\{i, j\} = \{x, \xi\}$  and  $\delta_{ij}$  is the Kronecker symbol.

The main quantity characterizing the kinetic properties of metals in a high frequency electromagnetic field is the surface impedance tensor

$$Z_{ij}(\omega) = -\frac{8i\omega}{c^2} \int_0^\infty dk D_{ij}^{-1}(\omega, k),$$

which relates the total current in the metal to the tangential components of the electric field at the sample surface, where the tensor  $D_{ij}$  is defined in Eq. (6) and  $D_{ij}^{-1}$  is the inverse of  $D_{ij}$ .



In order to obtain simple analytic expressions for the conductivity and surface impedance, we use a model for the electron energy spectrum that corresponds to the approximations of weak coupling in the plane of the layers and strong coupling for electrons belonging to adjacent layers

$$\varepsilon(\mathbf{p}) = \frac{p_x^2 + p_y^2}{2m} - \varepsilon_0 \cos \frac{p_z}{p_0}. \quad (7)$$

Here  $\varepsilon_0 \equiv \eta v_F p_0 \ll \varepsilon_F$  is the overlap integral for the electron wave functions,  $v_F^2 \equiv 2\varepsilon_F/m$ ,  $m$ , and  $m$  is the effective mass.

When  $\eta \tan \vartheta \ll 1$ , in the first approximation with respect to  $\eta$  the cyclotron frequency

$$\omega_H = (|e|H_0/mc) \cos \vartheta,$$

is independent of the projection  $p_H$  of the momentum in the magnetic field direction and the components of the electron velocity  $v_x^{(0)}, v_y^{(0)}$  are the trigonometric functions

$$v_x^{(0)}(t) = -v_F \sin(\omega_H t), \quad v_y^{(0)}(t) = v_F \cos(\omega_H t).$$

The velocity perpendicular to the layers is given by

$$v_z(t) = \eta v_F \sin\left(\frac{p_H}{p_0 \cos \vartheta} - \frac{mv_y(t)}{p_0} \tan \vartheta\right).$$

If  $\vartheta$  is close to  $\pi/2$ , then the closed intersections of the FS with the plane  $p_H = \text{const}$  are highly elongated and an electron cannot make a full turn in its orbit in momentum space over the mean free time. Thus, in the following we assume that  $\eta \tan \vartheta \ll 1$ .

When the skin effect is strong, i.e.,  $kv_F \sin \phi \gg \omega_H$ ,  $\omega$ , the integrals with respect to  $t$  and  $t_1$  in Eq. (5) can be calculated by the stationary phase method.<sup>19</sup> The stationary points are determined from the equations

$$v_x^{(0)}(t) = v_y^{(0)}(t) \sin \phi = 0, \quad v_x^{(0)}(t - t_1) = 0,$$

since  $kv_z \simeq \eta kv_F \ll kv_F$ . The largest component of the tensor  $\sigma_{ij}$  is  $\sigma_{xx}$ , which is proportional to  $(kr_0)^{-1}$ , where  $r_0 = v_F/\omega_H$ . The expansion in powers of the components  $\sigma_{yi}$ ,  $i = x, y, z$  begins with terms of a higher order in  $(kr_0)^{-1}$ .  $|\sigma_{xy}|^2$  is small compared to  $|\sigma_{xx}\sigma_{yy}|$ . The components  $\sigma_{zi}$  ( $i = x, y$ ) are proportional to  $\eta$ , and  $\sigma_{zz} \propto \eta^2$ . Therefore, in the case examined here the asymptote of the conductivity tensor  $\sigma_{ij}$  becomes diagonal.

In the first order approximation with respect to the small parameters  $(kr_0)^{-1}$  and  $\eta$ , the largest component of the conductivity tensor is given by

$$\sigma_{xx}(k) = \frac{i\omega_p^2}{4\pi^2\omega_H k_y r_0} \int_{-\pi}^{\pi} d\beta \frac{\cos \frac{\pi}{\omega_H}(\tilde{\omega} - \langle \mathbf{k}\mathbf{v} \rangle) - \sin R_0}{\sin \frac{\pi}{\omega_H}(\tilde{\omega} - \langle \mathbf{k}\mathbf{v} \rangle)}, \quad (8)$$

where  $\omega_p = \sqrt{4\pi n_0 e^2/m}$  is the plasma frequency,  $n_0$  is the electron density

$$R_0 = \omega_H^{-1} \int_{-\pi/2}^{\pi/2} d\phi \mathbf{k}\tilde{\mathbf{v}}(\phi) \approx 2k_y v_F/\omega_H = kd_0,$$

$\phi = \omega_H t$ ,  $d_0 = 2r_0 \sin \phi$  is the displacement of an electron along the  $\xi$  axis over a half period  $\pi/\omega_H$ ,  $\tilde{\mathbf{v}} = \mathbf{v} - \langle \mathbf{v} \rangle$ , and  $\beta = p_H/(p_0 \cos \vartheta)$ . The average  $\langle \mathbf{k}\mathbf{v} \rangle$  in the first approximation with respect to  $\eta$  is given by

$$\langle \mathbf{k}\mathbf{v} \rangle = \eta kv_F J_0(\alpha) (\sin \phi \tan \vartheta + \cos \phi) \sin \beta, \quad (9)$$

where  $\alpha = (mv_F/p_0) \tan \vartheta$  and  $J_0(\alpha)$  is the Bessel function.

In the collisionless limit  $\tau^{-1} \rightarrow 0$ , Eq. (8) can be written as

$$\sigma_{xx} = \frac{\omega_p^2}{\pi k d_0 \omega_H} \left\{ \sum_{n'} \frac{1 - (-1)^{n'} \sin(kd_0)}{\sqrt{[\eta k_1 v_F J_0(\alpha)]^2 (-\omega - n'\omega_H)^2}} + i \sum_{n''} \frac{\text{sgn}(\omega - n''\omega_H) [1 - (-1)^{n''} \sin(kd_0)]}{\sqrt{(\omega - n''\omega_H)^2 - [\eta k_1 v_F J_0(\alpha)]^2}} \right\}. \quad (10)$$

The sum in Eq. (10) is taken over  $n'$  and  $n''$  such that the inequalities

$$(\omega - n'\omega_H)^2 - \langle \mathbf{k}\mathbf{v} \rangle^2 < 0 \quad \text{and} \quad (\omega - n''\omega_H)^2 - \langle \mathbf{k}\mathbf{v} \rangle^2 > 0,$$

respectively, are satisfied and  $k_1 = k(\sin \phi \tan \vartheta + \cos \phi)$ . Terms with the same absolute magnitude and opposite sign in  $n''$  are assumed to be combined in a single term for absolute convergence of the series. The conductivity has sharp maxima when the resonance absorption condition (1) is satisfied. For the corresponding values of  $\omega$  and  $\mathbf{k}$  the impedance

$$Z_{xx} = -\frac{8i\omega}{c^2} \int_0^\infty \frac{dk}{k^2 - 4\pi i \omega c^{-2} \sigma_{xx}(k)} \equiv R - iX, \quad (11)$$

takes a minimum value. The real and imaginary parts of the impedance  $Z_{xx}$  are plotted as a function of  $\omega/\omega_H$  in Fig. 1 for  $\omega\tau = 20$  and different values of the anisotropy parameter  $\eta$ .

The model electron energy spectrum (7) can be used for a graphical representation of the resonance dependence of the surface impedance on the external magnetic field. For an arbitrary FS in the form of a slightly corrugated cylinder the shape of the resonance curves changes slightly, but the previous qualitative resonance behavior of the impedance is retained.

The characteristic feature of high frequency resonances in layered conductors is a small difference in the periods of the electron motion in different intersections of the FS with a plane  $p_H = \text{const}$ . Unlike ordinary metals, for which  $kv_D \gg \omega_H$ , in quasi-two-dimensional wires in a tilted magnetic field the oscillatory dependence of the impedance on  $H_0^{-1}$  shows up in the first approximation with respect to the parameter  $\delta/r_0$ . When  $kv_D \simeq \eta kv_F \simeq \omega_H$ , for resonance conditions the conductivity has a square root singularity, while the amplitude of the oscillations in the impedance increases as the anisotropy parameter of the FS becomes smaller. This latter circumstance is related to a reduction in the Landau damping. In conductors with small anisotropy parameters

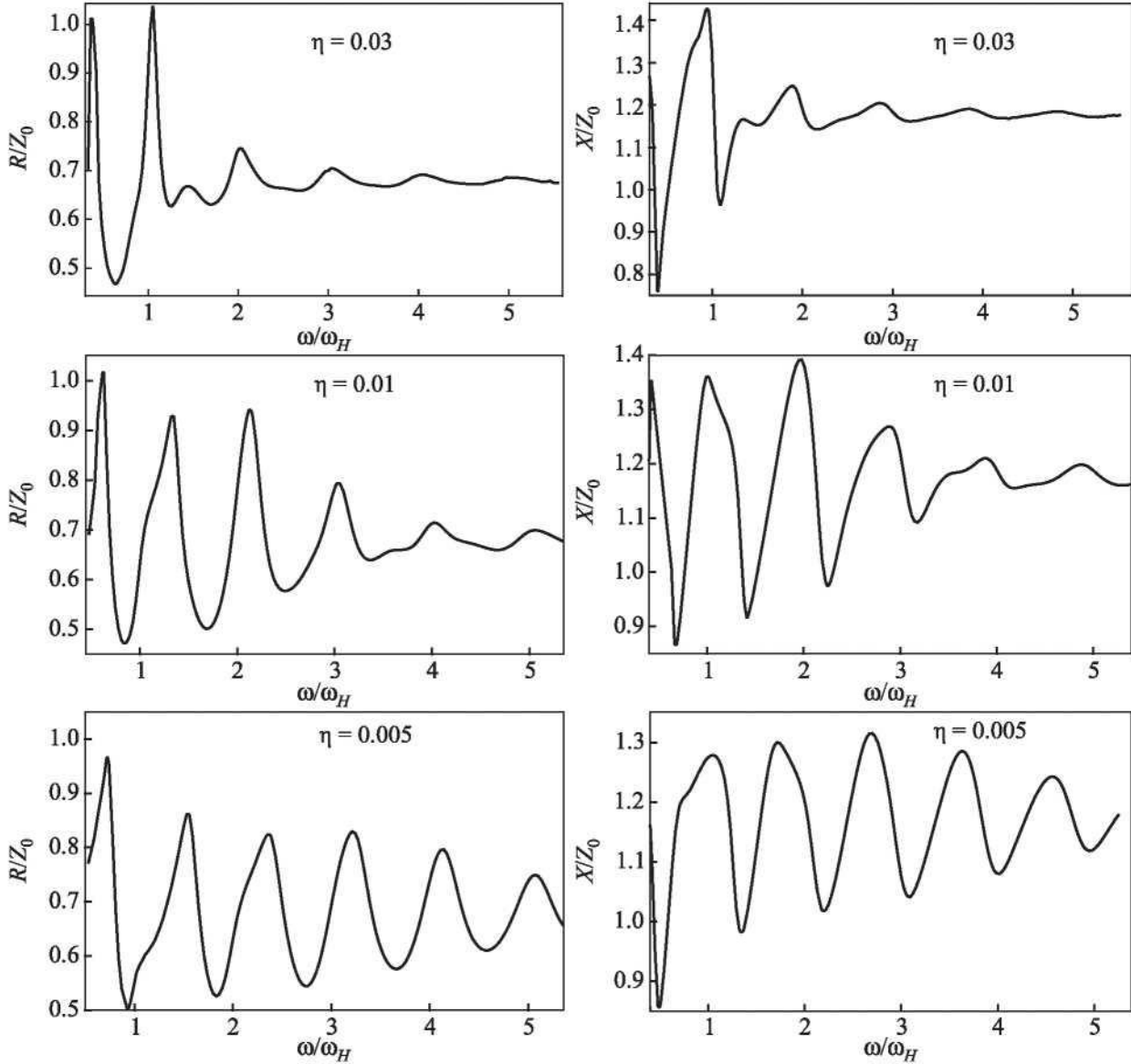


FIG. 1.  $R/Z_0$  and  $X/Z_0$  as functions of  $\omega/\omega_H$ , where  $Z_0 = 8\omega/c^2 k_0$  and  $k_0 = (2\omega_P^2 \omega / v_F c^2)^{1/2}$ , for  $\omega\tau = 20$ ,  $k_0 v_F / \omega = 50$ ,  $\vartheta = \pi/6$ ,  $\phi = \pi/4$ ,  $mv_F / p_0 = 2$ , and various values of  $\eta$ .

$\eta k v_F \ll \omega_H$ , for those directions of  $\mathbf{H}_0$  such that  $v_D$  is close to zero, there is no Landau absorption and even under conditions of a strong skin effect the cyclotron resonance has the same intensity as in a magnetic field parallel to the sample surface.

In this paper we have primarily studied organic metals in the family of tetrathiafulvalene salts. The conductivity in the plane of the layers,  $\sigma_{||}$ , for these materials is smaller than but comparable to the conductivity of ordinary metals, while the ratio of  $\sigma_{||}$  to the interlayer conductivity  $\sigma_{\perp}$  is usually on the order of  $10^3$ – $10^4$ . A simple estimate shows that the condition  $v_D T \simeq \eta v_F T \geq \delta$  for Landau damping to have a significant effect on the high-frequency absorption can easily be realized. Our results may also be valid for other layered structures with quasi-two-dimensional electron energy spectra, such as the dichalcogenides of the transition metals, cuprates, etc., which have a charge carrier density high enough to produce an anomalous skin effect at frequencies on the order of 100 GHz at low temperatures.

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