Surface pinning in amorphous ZrTiCuNiBe alloy

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We have measured the amplitude and the phase of an electromagnetic (EM) field radiated from superconductor (amorphous ZrTiCuNiBe alloy) in the mixed state due to interaction of the flux lattice with an elastic wave. The results undoubtedly point to an essential contribution of a surface pinning into the flux lattice dynamics. We propose a model that describes radiation of EM field from superconductors with non-uniform pinning. The model allows to reconstruct the viscosity and the Labush parameters from the experimental data. The behavior of the Labush parameter can be qualitatively explained in terms of the collective pinning theory with the allowance of thermal fluctuations.

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Soft type-II superconductors were studied intensively during last four decades with the goal to investigate various aspects of vortex matter dynamics (see the comprehensive review by Brandt ²). Nevertheless, many questions that require further investigation remain. Among these questions is the problem of relation between the surface and the bulk pinning. Up to now the dynamics of the vortex state in such an exclusively non-uniform situation was studied by the surface impedance method ². In this paper on the example of the amorphous Zr₄₁.₂Ti₃₃.₈Cu₁₂.₅Ni₁₀Be₂₂.₅ alloy we demonstrate the abilities of a new method based on excitation of the vortex lattice oscillations by a high-frequency sound wave.

The essence of the method is the following. A superconductor situated in lower half-space (z < 0) is subjected by a constant magnetic field H || z. A transverse elastic wave propagating along H and polarized in x direction produces transverse (with respect to H) oscillations of the vortex lattice caused by pinning forces and viscous friction forces, and, consequently, induces electromagnetic (EM) field. An antenna receives EM field (with Eᵢ and Hᵢ components) radiated through the elastically free surface of the sample (the surface perpendicular to the direction of propagation of the elastic wave). While similar experimental setup was already applied for the study of type-II superconductors ³, ⁴, the key new point is measuring both the amplitude and the phase of EM field (more accurately, the changes of these quantities).

In the uniform case and in the local limit (q ≫ l⁻¹, q is the wave number and l is the mean free path) the components of EM field at z = 0 are given by a simple expression ³, ⁴

\[ H_x = E_y = \frac{u(0)}{c} H \pi \frac{k^2}{q^2 + k^2}, \]  

where \( u(z) = u_0 \cos qz e^{i\omega t} \) is the elastic displacement, \( k^2 \) is the square of complex wave number of EM field in a conductor, and \( c \) is the light velocity.

Fig. 1: Amplitude and phase of EM field measured for the sample with imperfect surface. Inset - behavior of |E|/H near \( H_{c2} \) (1 - experiment, 2 - computed from Eq. (7) with \( \alpha_{L_{eff}} \) (see below))

Eq. (1) is applicable for normal as well as for superconducting state of metal. In normal state \( k^2 = k_n^2 = 4\pi \omega \sigma_0 / c^2 \) (\( \sigma_0 \) is the static conductivity in normal state) is imaginary valued quantity. In superconducting state at small magnetic fields \( (H \sim H_{c1}) \) \( k^2 = k_s^2 = \lambda_L^{-2} \) (\( \lambda_L \) is the London penetration length) is real valued. In well-developed Shubnikov state \( k^2 = k_m^2 = 4\pi (\omega \eta + \alpha_L) / H^2 \), where \( \eta > 0 \) is the viscosity parameter and \( \alpha_L > 0 \) is the Labusch “spring” parameter. We count the phase \( \varphi \) of EM field from its value at small \( (H = H_{c1} + 0) \) magnetic field. As follows from Eq. (1), the phase is positive at all \( H > H_{c1} \) and it approaches \( \varphi_n < 90^\circ \) (the phase in normal state) at \( H \rightarrow H_{c2} \).

The initial goal of our experiment was to obtain experimentally the dependencies \( \eta(H) \) and \( \alpha_L(H) \) using Eq. (1) as it was done before for MgB₂ ⁵. We use the working frequency \( \omega / 2\pi \sim 55 \text{ MHz} \). Details of the measuring procedure are described in ³. The results of our first experiments are presented in Fig. 1.

Prior discussing these results let us give the values of parameters for the material under study that important for further analysis. The measured d.c. resistiv-
unity $\rho \approx 200 \mu \Omega \cdot \text{cm}$ is practically temperature independent. The Alfer-Rubin effect (quadratic dependence of the sound attenuation coefficient on the magnetic field) allows to determine the parameter $\beta = |q^2/k_n^2| = 91 \pm 1$, in excellent coincidence with the value of $\rho$ measured (the sound velocity was found in [10]). The Hall constant, measured at room temperature, is positive and very small: $R_H = \frac{1}{\sigma_{||}} = (3.2 \pm 0.4) \cdot 10^{-25}$ CGS units, that yields rather high density of the carriers $n \approx 2.2 \cdot 10^{23} \text{cm}^{-3}$. Taking $m = m_e$ we obtain the relaxation time $\tau \approx 0.8 \cdot 10^{-16}\text{s}$ and for the Fermi velocity $v_F \approx 10^6\text{cm/s}$ we find $l \approx 10^{-8}\text{cm}$, which is close to the dielectricization threshold. It was shown before [11] that the alloy under study belongs to the family of weak coupling superconductors with the standard BSC energy gap $\Delta(0) \approx 1.75T_c$ ($T_c \approx 0.85\text{K}$). In the dirty limit the formula for the penetration depth [12] can be rewritten as $\lambda_T^2 = \frac{k_n^2}{\pi^2} \tanh \frac{T}{T_c}$, that yields $\lambda_T = 3 \cdot 10^{-4} \text{cm}$ at $T = 0.4\text{K}$. The coherence length calculated from $H_{c2}$ is $\xi(0.4\text{K}) = 1.4 \cdot 10^{-6}\text{cm}$ and the Ginzburg-Landau parameter is $\kappa = \lambda_L/\xi \approx 200$.

As follows from Eq. (1), for $\beta$ given above one could expect $\varphi_n \approx 90^\circ$ that contradicts with the result presented in Fig. 1 ($\varphi_n \approx 120^\circ$). Let us show that the discrepancy found can be accounted for lowering of conductivity near the surface of the sample caused by imperfectness of the surface.

We imply the following model dependence of the conductivity on $z$: $\sigma_0(z) = \sigma_0[1 - p \exp(z/z_\sigma)]$ (with $p \leq 1$ and $z_\sigma > 0$). Then in normal state the electrodynamic equation for the EM field has the form (see [4]):

\[
\frac{d^2\tilde{E}}{dz^2} - a(\zeta)\tilde{E} = a(\zeta)\cos(\zeta),
\tag{2}
\]

where $a(\zeta) = \alpha(1 - p \exp(z/z_\sigma)), a = k_n^2/q^2, k_n^2 = 4\pi\omega\sigma_0 z^2$.

Here we use the dimensionless variables $\tilde{E} = E/(i\omega_0 H)$ and $\zeta = qz$. The boundary conditions for Eq. (2) are that $|\tilde{E}(\zeta)|$ is finite at all $\zeta$ and $d\tilde{E}/d\zeta|_{\zeta=0} = 0$. The latter condition is due to continuity of EM field on the conductor-vacuum interface and is valid with the accuracy $\delta/\lambda_{EM} \lesssim 10^{-4}$ ($\delta$ is the skin depth and $\lambda_{EM}$ is the wavelength of EM field in vacuum).

The solution of Eq. (3) can be expressed through the Bessel functions of the 1-st kind of complex order $\nu = 2\zeta_0\sqrt{\alpha}$ on complex variable $t(\zeta) = 2\zeta_0\sqrt{\alpha}\exp(\zeta/2\zeta_0)$. The field at the conductor-vacuum interface is determined by the expression:

\[
\tilde{E}(z = 0) = \left(\frac{dJ_\nu[t(\zeta)]}{dz}\right)_{\zeta=0}^{-1} \int_{-\infty}^{0} dz J_\nu[t(\zeta)]a(\zeta)\cos(\zeta).
\tag{3}
\]

One can evaluate Eq. (3) using the expansion of $J_\nu(t)$ in series in $t$ and integrating each term analytically. For the case of interest it is enough to take into account first two terms of the expansion.

Analysis of Eq. (3) shows that the phase $\varphi_n$ increases under increasing $p$. In our case of extremely dirty conductor ($|\alpha| \sim 10^{-2}$) and for $\zeta_0 \sim 1$ we find that a rather small lowering of surface conductivity ($\sim 10\%$) comparing to the bulk one results in increasing of $\varphi_n$ up to $120^\circ$.

Removing the layer $\sim 50 \text{\mu m}$ from the surface by additional polishing with a fine powder (the size of grains $\sim 1 \div 2 \text{\mu m}$) we managed to eliminate the lowering of surface conductivity. The results of measurements (at various $T$) are shown in Fig. 2. One can see that in this case $\varphi_n \approx 90^\circ$. The main peculiarity of the dependencies observed is that at all temperatures the phase $\varphi$ becomes negative at intermediate magnetic fields. This effect cannot be described by Eq. (1) under any reasonable variation of $\eta$ and $\alpha_L$ with $H$.

In what follows we will argue that the effect observed can be explained by non-uniform pinning near the surface of the sample that takes place even in a situation with uniform conductivity (and, consequently, the uniform parameter $\eta$). Since peculiar behavior of the phase are observed in samples having less perfect as well as more perfect surface we consider the non-uniform pinning be an intrinsic property of the surface of the material under study.

EM field in the mixed state is described by the system of equations, namely, the Maxwell equation, the matter equation, that determines the value of the current in the two-fluid model, and the equation of motion for the vortex lattice [4]:

\[
\frac{d^2\mathbf{E}}{dz^2} = \frac{4\pi i\omega}{c^2} j = k_n^2(\mathbf{E} + \frac{1}{c}\mathbf{u}_v \times \mathbf{H}),
\tag{4}
\]

\[
\frac{1}{c} \mathbf{j} \times \mathbf{H} + i\omega(\mathbf{u}_v - \mathbf{u}_c) + \alpha_L(\mathbf{u}_v - \mathbf{u}_c) = 0,
\tag{5}
\]

where $\mathbf{u}_v$ is the displacement of the vortex lattice. Here
we neglect the normal component of the current.

Non-uniform pinning can be modelled by $z$-dependent Labush parameter. We specify this dependence as

$$\alpha_L(z) = \alpha_{L_{\perp}} + \alpha_{L_{z}} \exp(z/z_p).$$

For the magnetic field satisfied the inequality $k^2H^2 \gg |\omega \eta + \alpha_L|$ we obtain from Eq. (1) the equation for EM field that coincides in form with Eq. (2) with the same boundary conditions and the quantity $a(\zeta)$ defined as

$$a(\zeta) = \frac{i\omega \eta + \alpha_{L_{\perp}}}{q^2 H^2} + \frac{\alpha_{L_{z}}}{q^2 H^2} \exp \frac{\zeta}{\zeta_0} \equiv A + C \exp \frac{\zeta}{\zeta_0},$$

where $\zeta_0 = q z_p$. 

Taking into account first two terms in the expansion of the Bessel function we obtain from Eq. (3) the approximate expression

$$\tilde{E}(0) = \frac{\sqrt{A} \frac{\zeta_0}{\sqrt{A} + C \zeta_0}}{\sqrt{A} + C \zeta_0}.$$ 

Eq. (4) can be used to reconstruct the dependencies $\eta(H)$ and $\alpha_L(H)$ from the experimental data presented in Fig. 2 if the parameters $\zeta_0$ and $\alpha_{L{\perp}}/\alpha_{L{z}}$ are specified. These parameters are not known, but there are limitations on the choice of them. We require the functions $\eta(H)$ and $\alpha_L(H)$ to be smooth and real valued. Numerical analysis shows that these requirements satisfy for $\alpha_{L_{z}} \ll \alpha_{L_{\perp}}$ (it means that the bulk pinning can be neglected) and $\zeta_0 \lesssim 0.5$.

The dependencies $\alpha_{L{z}}(H)$ obtained for $\zeta_0 = 0.5$ are shown in Fig. 3.

There are physical reasons for $\zeta_0$ to be close to $\zeta_0 = 0.5$. One can see from Eq. (4) that smaller values of $\zeta_0$ correspond to larger values of $\alpha_{L{z}}$. But down to the lowest temperatures available in our experiment we do not find any signs of freezing of the magnetic flux (the irreversibility line). It means that the pinning is quite weak and it is caused, most probably, by point defects. The estimates (see further) based on the collective pinning (CP) theory show that the values of $\alpha_{L{z}}$ for $\zeta_0 = 0.5$ (presented in Fig. 3) are close to maximal possible value of this parameter.

It is interesting to note that $\zeta_0 \sim 0.5$ corresponds to $z_p \sim \lambda_L$. Probably, it is a fingerprint of that the image forces are responsible, in some part, for a formation of $z$-profile of the pinning potential.

The dependencies $\eta(H)$ are unsensitive to the choice of $\zeta_0$ and linear in a rather wide interval of $H$ with the slope $S \propto H_{c2}$ (Fig. 4). In this interval they have the form $\eta(H) = (2 \pm 0.05) H H_{c2} \sigma_n / c^2$

The CP theory allows to compute the field and temperature dependence of the Labush constant using only one free parameter. This parameter is connected with the quenched disorder of the vortex lattice caused by a random pinning potential. It is convenient to choose a field $H_{sv} \leq 0.5 H_{c2}$ as such a parameter [14]. This field separates the regime of single vortex pinning (SVP) from the regime of bundle vortex pinning (BVP). In the SVP regime ($H \leq H_{sv}$ or $H \geq H_{c2} - H_{sv}$), the value of $\alpha_L$ is given by the expression $\alpha_L \approx C_{66} a_v^{-2} (H_{sv})$, where $C_{66} = g_0 H_{c2} \left(1 - \frac{H_{c2}}{H_{c1}}\right)^2$ is the shear modulus of the vortex lattice, $a_v(H) = \sqrt{\Phi_0 / H}$ is the vortex lattice constant, and $\Phi_0$ is the quantum of magnetic flux. In the BVP regime ($H_{sv} \leq H \leq H_{c2} - H_{sv}$) the Labush parameter is $\alpha_L \approx C_{66} R_c^{-2} (H)$, where the collective pinning radius $R_c$ is given by the formula [18]

$$R_c(H) \approx a_v(H) \exp \left\{ \frac{1}{2} \left( \frac{H_{c2} - H}{H_{sv} (H_{c2} - H_{sv})} \right)^2 - 1 \right\},$$

The expressions presented determine the dependence $\alpha_L(H)$ almost symmetric relative to the $H = 0.5 H_{c2}$ line with the shape varied from the bell shape at $H_{sv} \sim 0.5 H_{c2}$ to the double-hump shape at $H_{sv} \ll H_{c2}$

One can see from Fig. 3 that while the value of $\alpha_L$ obtained from the experimental data is of the same order as one given by the CP theory estimates, but the dependence $\alpha_L(H)$ is strongly asymmetric relative to the $H = 0.5 H_{c2}$ line. We consider that such behavior of $\alpha_L$
are caused by thermal fluctuations.

To evaluate the effect of thermal fluctuations one should add a random force $f_L$ into Eq. (6). Strictly speaking, the returning force $\gamma H/c = (H^2/4\pi)\partial^2 u_{\phi}/\partial z^2$ in Eq. (5) has to be found from joint solution of the system (4), (5), but for the estimates one can replace $\partial^2/\partial z^2$ with $-q^2$. In this case Eq. (5) can be rewritten in the form of equation of diffusion of the Brownian particle

$$\eta \frac{\partial w}{\partial t} = \frac{\partial}{\partial w}(V_0 + V_u + V_p) + f_L,$$

where $w = u - u_c$, the "tildes" indicate that the corresponding quantities are given per one diffusing "particle": $\tilde{y} = y\Phi_0 L_x/H$ ($L_x$ is the collective pinning length), $V_0 = (q^2 H^2/4\pi)w^2$, $V_u = (q^2 H^2/4\pi)\omega^2$, and $V_p$ is the pinning potential. The latter is modelled by a three-well potential

$$V_p = \frac{\alpha_L}{2} \begin{cases} (w + d)^2 & w \leq -d/2 \\ w^2 & |w| \leq d/2 \\ (w - d)^2 & w \geq d/2 \end{cases}$$

where $d$ is the distance between minima of the pinning potential. The linearized Fokker-Planck equation that corresponds to Eq. (9) with the pinning potential (10) has the exact solution in terms of hypergeometric functions. At $\alpha_L/(q^2 H^2) \ll 1$ the following simple estimate for the averaged displacement of the vortex lattice is found $\langle w_{\phi} \rangle \approx \eta \cdot 4\pi (\omega^2 + c^2)/(q^2 B^2)$, where $c^2 = \frac{q^2 H \Phi_0 L_x}{2\pi^2} d^2$. One can see from the comparison of this formula with Fig. 3 that the thermal fluctuations may result in an essential reduction of the effective Labush parameter remaining the viscosity parameter unchanged. For general case the dependencies $\alpha_{eff}^c(H)$ obtained from the solution of the Fokker-Planck equation are shown in Fig. 3. To achieve semi-quantitative agreement between the theoretical results and the experimental data at $T = 0.4K$ we choose $H_{sc} \approx 0.15H_{c2}$ and $d \approx 5 \cdot 10^{-7} cm$. Other important points of the fitting procedure are the following: I) To agree the maximum value of $\alpha_{eff}^c$ (at 0.4K) with the experimental one we take for $\lambda_\phi$ that enter into equation for $C_{66}$ the value $\lambda_\phi(0.4K) = 1.2 \cdot 10^{-4} cm$. The standard temperature dependence of $\lambda_\phi$ is implied. II) We set $L_x = a_\phi(H)$. III) The quantity $d$ is assumed temperature dependent ($d(T) \propto \xi(T)$). IV) We imply that temperature dependence of $H_{sc}$ is determined by the $\delta T_c$ pinning: $H_{sc}/H_{c2} \propto (1 - (T/T_c)\delta)^{1/3}$. Such an assumption looks quite reasonable if one takes into account that there is a superconducting phase with higher $T_c$ on the surface of our sample. 

One can see from Fig. 3 that computed temperature and field dependencies of $\alpha_L$ are in qualitative agreement with the experimental results.

It is interesting to note that if we substitute $\alpha_{eff}^c$ into Eq. (7) we obtain $|E|/H$ that has a local minimum near $H_{c2}$. Such a minimum is observed sometimes in our experiment (Fig. 1) as well as in [3]. Physically this minimum is connected with the screening of EM field radiated from dipper regions of the sample by the surface layer with reduced penetration depth. Reminiscence of this minimum is also present in Fig. 2 under transition from the normal to mixed state the increase of $|E|$ begins only after substantial lowering of $\varphi$.

In conclusion, we propose the new method of investigation of dynamical characteristics of the vortex matter that consists in measuring the amplitude and the phase of EM field radiated from the sample under excitation of the vortex lattice oscillations by elastic wave. It is established that unusual behavior of the amplitude and the phase of EM field are accounted for the surface pinning. The parameters measured (viscosity coefficient and Labush parameter) are in good agreement with the estimates obtained from the theories of vortex matter dynamics.

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