

# Piezomagnetism of FeSe single crystals

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**Abstract** – The acoustic-electric transformation in high-quality FeSe single crystals is studied. In zero magnetic field we observe an abnormally strong electromagnetic radiation induced by a transverse elastic wave. Usually a radiation of such intensity and polarization is observed only in metals subjected to a high magnetic field (the radiation is caused by the Hall current). We argue that in FeSe in zero magnetic field it is caused by the piezomagnetic effect which is most probably of dynamical origin. We find that the piezomagnetism survives under the transition from the normal to superconducting state. In the superconducting state the electromagnetic signal decreases with decreasing temperature that is connected with the change in the London penetration depth.

An interplay between magnetic order and superconducting properties of 11-type Fe-based superconductors remains an open question. For the most part, it is connected with the fact that the magnetic state of such compounds is uncertain. For the first time, epitaxial films of tetragonal FeSe were studied at room temperature and they behaved as ferromagnets with saturation magnetization  $\sim 500$  emu/cm<sup>3</sup> [1]. In [1] it was also mentioned a manifestation of the anomalous Hall effect (AHE) in FeSe films. Unfortunately, any information on the superconducting properties of these films was not obtained. The AHE was also observed in epitaxial superconducting films FeSe<sub>1-x</sub>Te<sub>x</sub> ( $x = 0.5$ ) [2], but the nature of the effect (ferromagnetic or antiferromagnetic) has not been clarified. In the muon experiment [3] it was found that FeSe polycrystals acquire the static magnetic order (presumably antiferromagnetic) at enhanced ( $> 0.8$  GPa) pressure. The nuclear magnetic resonance experiment on a polycrystalline FeSe [4] indicates an increase in antiferromagnetic fluctuations close to the superconducting transition point. A weak ferromagnetism with saturation magnetization  $\sim 0.2$  emu/cm<sup>3</sup> was registered in FeSe single crystals [5]. At the same time, the neutron diffraction [3] and Mossbauer experiments [6] did not reveal any magnetic order in FeSe

polycrystals.

In all the papers cited above the samples were single-phase objects (as confirmed by the X-ray analysis) and there is no reason to attribute magnetic ordering (or its tracks) to possible impurity phases. One should agree with the point of view of [4] that FeSe is very close to the transition into a magnetically ordered state, and minor variations in the composition or internal stresses may favor the transition (or, on the contrary, suppress it). In this connection the study of perfect single crystals which composition provides a high degree of homogeneity becomes of great importance.

In this letter we present the results of experiments on an acoustic-electric transformation (AET) in high-quality FeSe<sub>0.963±0.005</sub> single crystals. These results are rather intriguing. In our opinion, they indicate the existence of the piezomagnetic (PZM) effect in this crystal. It is commonly known [7] that piezomagnetism is only possible in a magnetically ordered state. We argue that in our case it is the state with a dynamical magnetic order. In the superconducting state the AET signal decreases, but it should be accounted for the change in the London penetration depth. The piezomagnetic coefficient remains of the same value as in the normal state.

Let us briefly describe the theoretical grounds for the AET experiment. The experimental setup is shown in the inset of Fig. 1. A more detailed information can be found in [8–10]. A transverse elastic wave with the wave vector  $\mathbf{q} = (0, 0, q)$  and the displacement vector  $\mathbf{u} = (u, 0, 0)$  enters into the sample through a delay line. This wave can be considered as an alternating and spatially modulated ion current. It produces an electromagnetic (EM) field which forces free electrons to move to compensate the initial disturbance, in accordance with the Le Chatelier principle. For nonmagnetic samples the resultant current and the resultant electrical component of the EM field are small by the ratio of the electron mass to the ion mass (the inertial Stewart-Tolman effect). They are aligned along the  $x$ -axis. In the magnetic field  $\mathbf{H} = (0, 0, H)$  the Lorentz force applied to free electrons results in the appearance of the  $y$ -components of the current and the electrical field (the Hall components). In view of lack of the compensating forces the Hall components exceed considerable the inertial ones. Due to continuity of the tangential components of the electric and magnetic field at the sample boundary the EM field is radiated from the surface. The radiation is registered by a polarized antenna (a flat coil). The antenna registers the magnetic component of the EM field (its projection on the normal to the antenna plane). Near the interface the EM field is the plane wave and its electrical component is equal the magnetic component. We will discuss later only the electrical component  $\mathbf{E}$ . Its projection on the antenna plane is referred to as the AET signal  $E$ . It is the complex quantity and in our experiment we measure its amplitude  $|E|$  and phase  $\arg(E)$  at different orientations of the antenna.

The EM field satisfies the Maxwell's equations that yield

$$\frac{d^2 \mathbf{E}}{dz^2} = \frac{4\pi i\omega}{c^2} \mathbf{j} + \frac{4\pi i\omega}{c} \nabla \times \mathbf{m}, \quad (1)$$

where  $\mathbf{j}$  is the resultant current, and  $\mathbf{m}$  is the magnetic moment induced by the elastic wave. The time dependence  $\propto \exp(i\omega t)$  is implied. For a nonmagnetic metal one can use the following local matter equation for the current

$$\mathbf{j} = \hat{\sigma}(\mathbf{E} + \mathbf{W}),$$

where  $\hat{\sigma}$  is the conductivity tensor the explicit form of which is well known from the theory of galvanomagnetic phenomena [11]

$$\hat{\sigma} = \sigma_0 \begin{pmatrix} 1 & -\Omega\tau \\ \Omega\tau & 1 \end{pmatrix}^{-1}$$

( $\sigma_0$  is the static conductivity,  $\tau$  is the relaxation time, and  $\Omega$  is the cyclotron frequency), and  $\mathbf{W} = (U_{ST}, U_{ind}, 0)$  is the extraneous electromotive force [12] with  $U_{ST} = (m/e)\omega^2 u$ , the Stewart-Tolman field, and  $U_{ind} = \frac{i\omega}{c} Bu$ , the inductive field ( $B$  is the magnetic induction in the sample).

For  $\mathbf{m} = 0$  in the limit  $\Omega\tau \ll 1$  the solution of eq. (1) has the form [10]

$$\begin{pmatrix} E_x \\ E_y \end{pmatrix} = -\frac{k_0^2}{q^2 + k_0^2} \begin{pmatrix} U_{ST} - \Omega\tau \frac{q^2}{q^2 + k_0^2} U_{ind} \\ U_{ind} \end{pmatrix}, \quad (2)$$

where  $k_0^2 = 4\pi i\omega\sigma_0/c^2$  is the square of the characteristic skin wave number. In deriving (2) we neglect the contribution of nonlocal effects (proportional to the electron mean free pass, see [8, 10, 12]) that are inessential for our analysis.

For a superconductor in the Meissner state ( $B = 0$ ) the current satisfies the London equation  $i\omega \mathbf{j} = (c^2/4\pi\lambda_L^2)(\mathbf{E} + \mathbf{W})$ , where  $\lambda_L$  is the London penetration depth. It yields

$$E_x = -\frac{k_s^2}{q^2 + k_s^2} U_{ST}, \quad E_y = 0, \quad (3)$$

where  $k_s^2 = \lambda_L^{-2}$ .

Thus in zero magnetic field ( $H = 0$ ) only the  $E_x$  projection of the electric field should be different from zero. Strictly speaking this conclusion corresponds to the case where all the quantities depend only on the  $z$ -coordinate (the one-dimensional case). The actual situation is a three-dimensional one, but if the transverse size of the sample exceeds the sound beam diameter the problem is described satisfactorily as an effectively one-dimensional one. In the latter case the dependence of the amplitude of the AET signal on the antenna orientation (the polarization diagram) demonstrates an almost 100 percent modulation. In smaller samples the distortion of the lines of the current near the side surfaces results in the appearance of the  $E_y$  component and in a reduction of the modulation index, but the maximum keeps at the same orientation as for large samples.

The measurements of the amplitude and phase of the AET signal can be considered as a powerful tool for the study of the vortex dynamics in superconductors. It allows to obtain quantitative information on the vortex viscosity, the vortex pinning strength [9], and the Magnus force [10], and even to estimate the vortex mass [13].

Our initial goal of the AET experiments with FeSe was the study of the dynamics of the vortex matter in this compound. The single crystals of FeSe were grown by the technology, described in [14]. The samples under investigation were the (001) facet platelets  $\sim 1.5 \times 1.5$  mm<sup>2</sup> in area. These samples were used previously for the study of acoustic characteristics [15]. The high quality of single crystals were confirmed by indexing the X-ray diffraction patterns [14] and by the observation of the  $\lambda$ -anomaly in the heat capacity at the superconducting transition [16]. The narrowness of the superconducting transition ( $\sim 0.5$ K) seen from the width of the longitudinal sound velocity jump [15] also witnesses for the sample quality. The measurements have been done in the pulsed mode at the frequency  $\sim 55$  MHz. The pulse power and duration is  $10 \div 30$  W/cm<sup>2</sup> and  $0.5$   $\mu$ s, correspondingly. The wave vector was directed

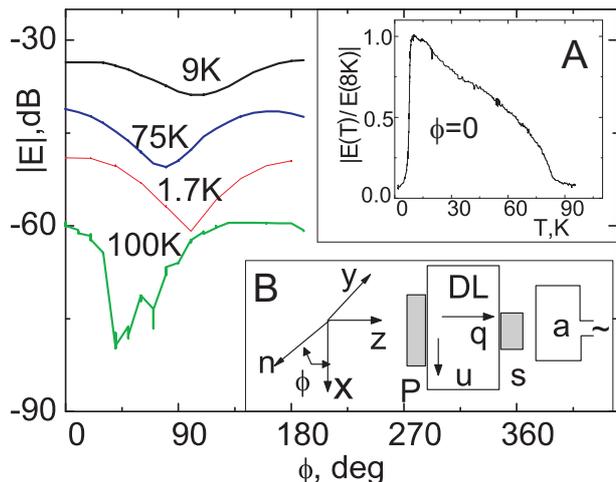


Fig. 1: The amplitude of the AET signals  $|E|$  versus  $\phi$ , the angle between the direction of the elastic displacement vector and the normal to the plane of the receiving antenna. The inset A is the temperature dependence of the amplitude at  $\phi = 0$ . The inset B is the experimental setup. Here P is the piezoelectric transducer, s is the sample, DL is the delay line, a is the polarized antenna, and  $\mathbf{n}$  is the normal to the antenna plane.

along the [001] axis of the sample. The diameter of the sound beam was  $\sim 3$  mm. We use the equipment described in [17]. To minimize thermoelastic stresses that emerge at the junction between the sample and the delay line a mylar film is embedded into the junction.

Against all the expectations, in the absence of an external magnetic field the recorded AET signal is in two to three order larger than one expected for the inertial field, and the maximum amplitude of the AET signal corresponds to the electrical field  $\mathbf{E}$  presumably polarized in the  $y$  (not  $x$ ) direction. The polarization diagrams for several temperatures are shown in Fig. 1. As is expected for small samples the modulation index is relatively small. The temperature dependence of the amplitude of AET signal in the  $y$ -direction ( $\phi = 0$ ) is shown in the inset A of Fig. 1. The signal appears below the temperature of solidification of the silicone oil (GKZH-94) used as a bonding material. Above the superconducting transition the amplitude of the signal grows monotonically under decrease in temperature. The temperature dependence has a jump in the derivative at the point of the tetra-ortho transformation in FeSe. In the superconducting state the amplitude of the signal decreases.

The magnetic field dependence demonstrates remarkable peculiarities. For  $\phi$  close to zero the dependence of the amplitude  $|E|$  on  $H$  has a deep minimum. At  $H$  that corresponds to the minimum of  $|E|$  the change in the phase of  $\mathbf{E}$  is close to  $\pi$ . Such behavior can be described in the vector diagram language. The AET signal behaves as a vector sum of two almost collinear components, an even in the magnetic field and an odd one:

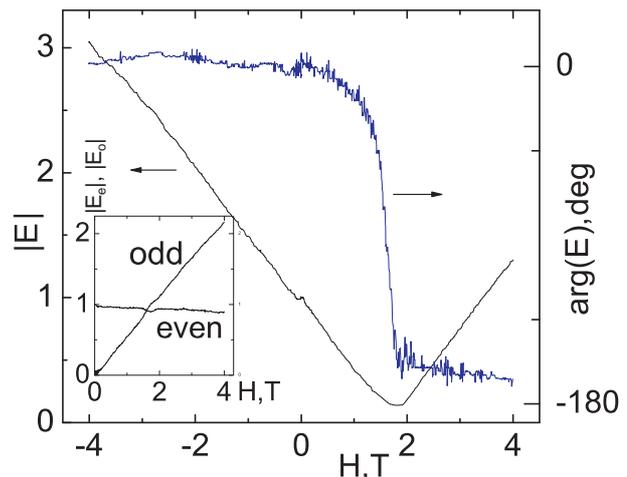


Fig. 2: The field dependence of the amplitude and phase of the AET signal at  $T = 9$  K and  $\phi = 20^\circ$ . The signal at  $H_0 = 0$  is used as the reference point for the phase and the scale for the amplitude. The inset shows the amplitudes of the even and odd components.

$E_{e(o)} = [E(H) \pm E(-H)]/2$ . The amplitudes  $|E_e|$  and  $|E_o|$  are shown in the inset of Fig. 2. One can see that the odd component is linear in the magnetic field and it is just the usual Hall component, while the even one is practically independent of  $H$ . The amplitude of the even component is equal to the amplitude of the odd (Hall) one at  $B \approx 2T$ . It allows us to estimate the value of the AET signal at zero magnetic field. It is in the factor of  $\Omega_{B=2T}/\omega$  larger than one caused by the inertial Stewart-Tolman force. For our frequency  $\Omega_{B=2T}/\omega \sim 10^3$ .

Fig. 2 shows the most demonstrative example where two components are in-phase or in antiphase. Under rotation of the antenna the amplitudes and phases of two components vary in different ways. Then, the position of the minimum is shifted and its depth is changed. In principle, it is possible to choose the antenna orientation for which the even and odd component are orthogonal to each other. In that case the minimum disappears. The position of the minimum depends on the quality of the surface treatment. In case of crude treatment the minimum moves to the range of fields inaccessible in our experiments ( $> 5.5T$ ). But in any case, the even and odd components demonstrate a behavior similar to one shown in the inset of Fig. 2, only the relation between their modules is changed. Let us also emphasize that we do not observe any hysteresis in the field dependencies (shown in Fig. 2 as well as obtained in other measurements), any nonlinear dependence of the signal on the sound amplitude, and any step-like features (that would be considered as a hallmark of spin-flop transitions).

Two effects may provide the Hall current in the absence of the magnetic field. They are the AHE and PZM effect and both of them are realized in magnetically ordered me-

dia. As a rule the AHE is observed in ferromagnetic conductors (see [18] and references therein), but, in principle, similar phenomenon is also possible in antiferromagnets (AFM) [19,20]. The intensity of the AHE can be characterized by the ratio of the off-diagonal component of the conductivity tensor to the diagonal one:  $\eta = \sigma_{xy}/\sigma_{xx}$ . This ratio yields the fraction of the transport current that branches off in a direction perpendicular to it. There are many experiments on the AHE in ferromagnets [18]. Experimental data indicate that the maximum value of  $\eta$  does not exceed  $10^{-2}$ . Unfortunately, there is no experimental statistics on the AHE in AFM. As far as we know the AHE was observed only in hematite [19]. For this compound the branch off factor is  $\eta \approx 2 \times 10^{-4}$ .

The ferromagnetic AHE implies that the sample has a spontaneous static magnetic moment  $M$ . Our attempt to register the magnetic field outside the samples was unsuccessful (we used the fluxgate sensor with sensitivity about 1 Oe and examined one sample with dimension  $1.4 \times 1 \times 0.3$ mm that corresponds to the demagnetizing factor  $\approx 0.7$ ). This means that even of the spontaneous moment exists, it is small enough, or it is concentrated in a thin surface layer that results in the effective demagnetizing factor close to unity. In the latter case the thickness of the magnetic layer is of order of the exchange force range  $a_0 \sim 10^{-7}$  cm.

Let us check whether the AHE with physically relevant  $\eta$  explains the observed anomaly. We write eq. (1) for the  $E_y$  component disregarding the contribution of the second term in the right hand side of eq. (1) and separating explicitly the anomalous part of the current

$$\frac{d^2 E_y}{dz^2} = \frac{4\pi i\omega}{c^2} (\sigma_0 E_y + j_{an}) = k_1^2 E_y, \quad (4)$$

where  $k_1^2 \approx -q^2$  and  $k_1^2 \approx a_0^{-2}$  in case of bulk and surface ferromagnetism, correspondingly, and

$$j_{an} = \eta(i\omega neu) \frac{M}{M_{max}} \quad (5)$$

is the anomalous current (here  $M_{max}$  is the saturation magnetic moment). The factor in the round brackets in eq. (5) corresponds to the  $x$ -component of the electron current caused by the sound beam (the analog of the transport current). A possible polydomain magnetic structure is accounted by the factor  $M/M_{max}$ . For an AMF  $k_1^2 \approx -q^2$  and the expression for the current (5) contains the antiferromagnetic vector  $L$  instead of  $M$ .

The quantity  $\eta$  can be evaluated taking into account that at  $B = B_0 \approx 2$  T the observed anomalous signal is of the same value as the usual Hall signal (Fig 2). From Eqs. (2), (4) and (5) we obtain

$$\eta = \left| \frac{k_1^2 - k_0^2}{q^2 + k_0^2} \right| \frac{\sigma_0 B_0}{nec} \frac{M_{max}}{M}. \quad (6)$$

The resistivity of our samples at  $T = 10$  K is  $\rho \approx 40 \mu\Omega \cdot \text{cm}$  [21] that yields  $|k_0^2| \approx 10^5 \text{ cm}^{-2}$ . The quantity  $q^2$  evaluated from the velocity of the  $C_{44}$  mode ( $s = 1.38 \times 10^5$

cm/s [15]) is  $q^2 = 6.25 \times 10^6 \text{ cm}^{-2}$ . The Hall measurements yield the density of carriers  $n \sim 10^{20} \div 10^{21} \text{ cm}^{-3}$  [1], but since the electron structure of FeSe corresponds to the compensated metal [22], this quantity is most likely overestimated. A more realistic estimate for  $n$  can be obtained from the London penetration depth: at  $T = 0$   $\lambda_L^2(0) = mc^2/4\pi ne^2$  [11]. For the FeSe<sub>0.94</sub> polycrystal the in-plane magnetic penetration depth  $\lambda_L(0) \approx 0.4 \div 0.5 \mu\text{m}$  was obtained in [23]. From this value, assuming that  $m$  is close to the free electron mass, we evaluate  $n \sim 10^{20} \text{ cm}^{-3}$ . This yields the branch-off factor  $\eta \sim 0.3$  (we put the factor  $M/M_{max}$  of order of unity). For our samples we obtain even larger  $\lambda_L(0)$  (see below) that corresponds to  $n \sim 10^{19} \text{ cm}^{-3}$  and  $\eta > 1$ . Assuming surface ferromagnetism we get  $\eta \gg 1$ . Such values of  $\eta$  are unphysical and we conclude that the anomalous AET behavior observed in our experiment cannot be accounted for the AHE.

Let us now take into account the PZM effect. Neglecting  $j_{an}$  and considering the case  $H = 0$  we obtain from eq. (1)

$$\frac{d^2 E_y}{dz^2} = k^2 E_y - \frac{4\pi i\omega}{c} \frac{dm_x}{dz}, \quad (7)$$

where  $k^2 = k_0^2$  and  $k^2 = k_s^2$  for the normal and superconducting state, correspondingly. In eq. (7)  $m_x$  is the  $x$ -component of the magnetic moment induced by the elastic wave. It can be presented as  $m_x = \sum_i \Pi_{xixz} L_i C_{44} du/dz$  [20], where  $C_{44}$  is the elastic constant,  $L_i$  is the  $i$ -th component of the vector of antiferromagnetism, and  $\Pi_{xixz}$  are the PZM coefficients (in [7,24] the PZM modulus  $\Lambda_0$  defined as  $\Lambda_0 = \sum_i \Pi_{xixz} L_i$  was used). The general form for the matrices of the PZM coefficients for different magnetic structures is given in [20].

Replacing  $d^2/dz^2$  with  $-q^2$ , we get

$$|E_y| = \frac{4\pi\omega}{c} \frac{q^2}{q^2 + k^2} \Lambda_0 C_{44} u. \quad (8)$$

For FeSe  $C_{44} \approx 10^{11} \text{ dyn/cm}^2$  [15]. From the condition that the Hall signal at  $B \approx 2$  T coincides with the PZM signal we obtain  $\Lambda_0 = 2.4 \times 10^{-10} \text{ emu}/(\text{dyn}\cdot\text{cm})$ . This is quite reasonable estimate that is one order less than the maximum known value for the PZM modulus (measured in CoF<sub>2</sub> [24]).

We would note that for the displacement vector polarized in the  $x$ -direction the appearance of the  $m_y$  component is not principally prohibited, especially in polydomain samples. In the latter case the PZM effect would be responsible for the appearance of the  $E_x$  component of the AET signal as well. In principle, it may explain the temperature shift of the extremum in the polarization diagrams.

The PZM effect considered is connected with that the elastic deformations violate the strictly antiparallel configuration of spins in AMF. In principle, another scenario that mimics the PZM effect can be realized. Let us imagine that the sample has a nonzero magnetic moment  $\mathbf{M}$  which direction is bound to the [001] axis by the anisotropy

forces. The origin of the magnetic moment can be the interstitial Fe or a magnetic phase grown at the surface (while each time prior to the cooling the working surface of the sample was cleaned by grinding with a fine abrasive powder, it does not exclude the presence of another phase of the atomic thickness at the surface). The elastic displacement  $\mathbf{u}$  produces a tilt of the [001] axis at the angle  $\varphi = \text{rot}\mathbf{u}/2$  [7] with respect to the  $z$  axis. Then the  $m_x = M(du/dz)/2$  component appears. The measured value of  $m_x$  is  $m_x = \Lambda_0 C_{44}(du/dz)$ , where  $\Lambda_0$  is given above. If the interstitial Fe were responsible for the effect observed, the bulk magnetization would be  $M = 2\Lambda_0 C_{44} \approx 50 \text{ emu/cm}^3$ . Then, the magnetic field near the sample surface is evaluated as  $B_z = 4\pi(1-b)M \approx 180 \text{ G}$  [7] ( $b = 0.7$  is the demagnetization factor). Such a field had to be registered easily by the fluxgate sensor, but this was not the case. To provide the same AET signal the magnetization of the magnetic phase at the surface would be by the factor of  $(qa_0)^{-1} > 10^3$  larger (per unit volume) than the bulk one. The latter possibility also looks unrealistic.

The explanation of our results in terms of the PZM effect immediately raises the question on the type of the magnetic structure in FeSe single crystals. In any case, it is obviously not a static AFM. It was shown in [24] that for the usual AFM the dependence of the PZM modulus on temperature reproduces the temperature dependence of the sublattice magnetization. It follows from (8) that at  $k_0^2 \ll q^2$  the behavior of  $|E_y|$  is determined by the behavior of the PZM modulus. One can see from Fig. 1 that in the normal state  $|E_y|$  increases almost linearly with the decrease of temperature, and it does not show any tendency to the saturation. It is most likely that in case of FeSe we deal with a dynamic short-range magnetic order. This conclusion correlates with an increase in AFM fluctuations in FeSe under approaching  $T_c$  [4] and justifies the negative results of the Mossbauer [6], neutron and  $\mu\text{SR}$  experiments [3]. But we would like to emphasize that for the observation of the PZM effect AFM fluctuations should have some preferable orientation of the antiferromagnetic vector.

The behavior of the AET signal in the superconducting state at  $H = 0$  is shown in Fig. 3. The signal decreases below  $T_c$ , but it does not disappear completely and saturates at the value easily accessible for the measurements. Taking into account that for the normal state  $k^2 = k_0^2 \ll q^2$  and for the superconduction state  $k^2 = k_s^2 = \lambda_L^{-2}$  we get from eq. (8) the relation

$$\frac{E_y^n(T_c)}{E_y^s(T)} = \left(1 + \frac{1}{q^2 \lambda_L^2(T)}\right) f(T). \quad (9)$$

Here the factor  $f(T)$  accounts possible changes in the PZM modulus in the superconducting state. Assuming  $f(T) = 1$  we obtain the temperature dependence  $\lambda_L^{-2}(T)$  shown in the inset of Fig. 3. The value  $\lambda_L(0)$  obtained as the limit of  $\lambda_L(T)$  at  $T \rightarrow 0$  is  $1.82 \pm 0.03 \mu\text{m}$ . This quantity can

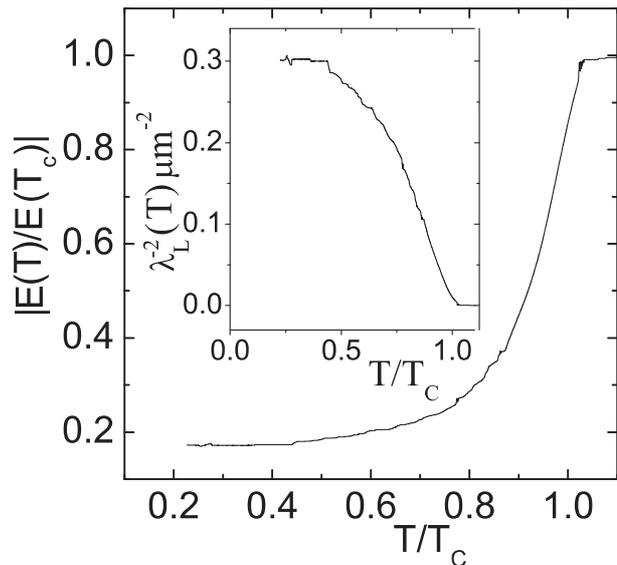


Fig. 3: The AET signal in the superconducting phase at  $\phi = 0$ . The inset shows the inverse square of the London penetration depth.

be also extracted from the slope of  $\lambda_L^{-2}(T)$  near  $T_c$  [11]:

$$\lambda_L^{-2}(T) = \lambda_L^{-2}(0) \frac{2(T_c - T)}{T_c} \quad (10)$$

that yields  $\lambda_L(0) = (1.65 \pm 0.1) \mu\text{m}$ . These estimates almost coincide to each other. This coincidence justifies our assumption that the PZM modulus remains unchanged below  $T_c$ . In other words, superconductivity and piezomagnetism co-exist "peacefully". One can in principle think about some phase separation scenario as a possible explanation of the behavior of the AET signal in the superconducting state. Then neither expression (9) nor (10) can be used for obtaining  $\lambda_L(0)$ , and  $\lambda_L(0)$  given by eq. (9) can coincide with the one by eq. (10) only accidentally. Therefore, the phase separation scenario looks doubtful.

We obtain  $\lambda_L(0)$  that is in three times larger than one found in [23] for the FeSe<sub>0.94</sub> polycrystal, and our estimate for  $\lambda_L(0)$  corresponds to the carrier density  $n \sim 10^{19} \text{ cm}^{-3}$ . Perhaps the discrepancy with [23] can be accounted by for a strong dependence of the carrier density on the structure of the sample and on its composition.

In summary, we have investigated the acoustoelectric transformation in high quality single crystals of FeSe. An abnormally strong electromagnetic radiation stimulated by the sound wave was detected in zero magnetic field. Most likely the nature of the effect is connected with the piezomagnetic properties of FeSe crystals. This implies that FeSe has some kind of magnetic order most probably the dynamical one. The value of the piezomagnetic constant is estimated. In the superconducting state the lowering of the AET signal can be accounted completely for by the change in the London penetration depth  $\lambda_L(T)$ . The latter means that the piezomagnetic interaction remains

unchanged. Our experiment yields a rather large estimate for  $\lambda_L(0)$ , that is apparently due to the low electron density.

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