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[†] In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.

Impact of spin correlations on resistivity and microwave absorption of $\mathrm{Ba}(\mathrm{Fe}_{1-x}\mathrm{Co}_x)_2\mathrm{As}_2^{\dagger}$

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The results of studies of BaFe₂As₂ single crystals doped with cobalt by means of resistivity and microwave absorption measurement are reported. A theoretical description of the behavior of the microwave absorption amplitude is made taking into account the temperature dependence of resistivity, magnetic susceptibility and the lifetime of spin fluctuations. An assumption has been made that the deviation from the linear dependence of resistivity on temperature at T < 100 K is not related to the electron-electron scattering mechanism, but it is due to the appearance of nematic fluctuations. Estimates of the rate of scattering by spin fluctuations indicate their nematic nature at temperatures near the structural transition.

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1. Introduction

Spin correlations are mostly considered as the main candidate for the role of mediator between free electrons when they combine into Cooper pairs upon emerging a superconducting state in iron based superconductors, in particular in the iron arsenide compounds. An essential reason for this statement is the fact that they determine the behavior of many physical parameters, in particular, the temperature dependence of resistance in the range above the superconducting transition, $T > T_c$ (see, f.e. [1-4]). In several theoretical papers [5-8], it was shown that the experimentally observed linear dependence of resistivity on temperature $\rho(T) \propto T$ (instead of the quadratic one characteristic of the scattering of quasiparticles in a Fermi liquid $\rho(T) \propto T^2$ is a consequence of their scattering just by spin fluctuations. Since antiferromagnetic spin correlations have a highly anisotropic order parameter corresponding to ordering in the form of spin density waves (SDW), they stimulate the establishment of nematic order in the temperature region near the transition point to the SDW phase T_N and nematic fluctuations above it (see Ref. [9] and references there). The latter manifest themselves both in magnetic measurements and in electronic conductivity in a wide temperature range, which extends to tens of degrees Kelvin above T_N . There is a statement that nematic fluctuations serve as a trigger for a structural transition from the symmetrical tetragonal crystal structure of C_4 to the anisotropic orthorhombic one with the C_2 symmetry [10, 11]. This transition occurs with a decrease in temperature at $T = T_s$, which is several degrees higher than the magnetic transition T_N .

Moreover, some authors point out the connection between transport properties (in particular, the contribution of spin fluctuations to the rate of carrier scattering) and superconducting parameters, such as the critical temperature T_c and the symmetry of the order parameter [3]. When studying the influence of different contributions to the scattering process, it is impor-

[†]This paper is dedicated to Professor Boris I. Kochelaev on the occasion of his 90th birthday.

tant to estimate and compare the scattering rates at centers of different types: defects $\tau_{\rm d}^{-1}$, impurities $\tau_{\rm imp}^{-1}$, phonons $\tau_{\rm ph}^{-1}$, spin fluctuations $\tau_{\rm sf}^{-1}$, etc. The value of the total rate $\tau_{\rm total}^{-1}$ can be determined using the results of measuring the resistance of the material to direct current. And to evaluate the spin-fluctuation contribution, it is necessary to study the response to an alternating electromagnetic field, which changes with a frequency ω comparable to the rate of scattering by spin fluctuations $\tau_{\rm sf}^{-1}$. For this purpose, in present work we used the method of recording microwave absorption at a frequency of $\sim 10^{10}$ Hz.

2. Experimental methods and technique

According to the classical Drude's theory, the resistivity of a material with metallic conductivity is determined by a small set of its physical parameters:

$$\rho = \frac{m^*}{e^2 n} \tau^{-1},\tag{1}$$

where m^* , e and n are an effective mass, charge and concentration of current carriers (electrons or holes). The scattering rates τ^{-1} consists of several contributions (as noted above): (i) scattering by defects and impurities τ_d^{-1} ; (ii) an "electron-electron scattering" of Fermi liquid quasiparticles upon interaction (collision) with each other τ_e^{-1} ; (iii) scattering by magnetic (spin) fluctuations τ_{sf}^{-1} , if the material has magnetic moments interacting with each other. The presence of other scattering processes is also possible, but in the compounds we studied, only those listed above act. Since the dependence of resistivity on temperature is determined mainly by changes in scattering rates, it can be described by the sum of three terms: $\rho(T) = \rho_o + \rho_e(T) + \rho_{sf}(T)$. The first term ρ_o , which due to scattering by impurities and defects, does not depend on temperature and determines the residual resistivity at T = 0. The temperature dependences of last two terms, ρ_e (due to electron-electron scattering) and ρ_{sf} (due to spin-fluctuation scattering), have different forms: $\rho_e(T) \propto T^2$ [12], while $\rho_{sf}(T) \propto T$ [5, 7, 8]. In general, the temperature dependence of resistivity has a form:

$$\rho(T) = \rho_0 + A_1 T + A_2 T^2.$$
(2)

Thus, by analyzing the experimental temperature dependence of the resistivity of the sample under study, it is possible to find out which scattering mechanism is predominant. Moreover, knowing the effective mass and carrier concentration, an estimate of the total dissipation rate τ^{-1} can be made using Equation (1).

In present study the character of the temperature dependence of electrical resistivity $\rho(T)$ was determined from the results of the R(T) measurement in the range from 4 to 300 K. Resistance measurements were carried out using a standard four-probe method at a constant current of about 1 mA magnitude. Current and potential contacts were attached using silver conductive paste on the *ab* plane of the crystal. In this way, the longitudinal component of the resistance R_{ab} was measured, which was then converted into resistivity ρ taking into account the dimensions of crystal.

To record microwave absorption (MWA), we used a standard BER-418s EPR spectrometer from Bruker, operating at a frequency of about 9.5 GHz. The amplitude of microwave losses A_{mwa} in the spectrometer cavity with a sample was measured in the temperature range from 4.2 K to 200 K.

It is known that in a conductive material the absorption of microwaves occurs in the skin layer. Therefore, the amplitude of the MWA is proportional to the volume of the skin layer, provided that the skin-depth δ is much greater than the electron mean free path $l_{\rm e}$ [13]. Consequently,

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a change in the skin-depth δ with decreasing temperature results in a temperature dependence of the amplitude of the microwave absorption signal A_{mwa} . Since the skin-depth is determined by the resistivity ρ through the equation $\delta = c \sqrt{\frac{\rho}{2\pi\omega\mu_0}}$ (here c is the speed of light, ω is the frequency and μ_0 is the permeability of vacuum), then it turns out that $A_{\text{mwa}} \propto \sqrt{\rho}$. This refers to the contribution of ohmic losses to microwave absorption. In addition, the MWA amplitude is affected by magnetic losses, which are proportional to the static magnetic susceptibility χ_0 . Their contribution depends on the ratio of the measurement frequency ω and the spin scattering rate, which is approximately equal to τ_{sf}^{-1} . Taking all this into account, the microwave absorption amplitude takes the form:

$$A_{\rm mwa}(T) = \sqrt{\frac{\mu_0 \omega_0}{2}} \sqrt{\rho(T)} \left(1 + \frac{1}{2} (\chi'(T) - \chi''(T)) \right), \tag{3}$$

with real χ' and imaginary χ'' components of magnetic susceptibility, which are expressed through the formulas:

$$\chi' = \chi_0 \frac{1}{1 + (\omega \tau_{\rm sf})^2},\tag{4}$$

$$\chi'' = \chi_0 \frac{\omega \tau_{\rm sf}}{1 + (\omega \tau_{\rm sf})^2}.$$
(5)

The Equation (3) was obtained by analogy with what S. Barnes did to describe the spin resonance of metal samples with magnetic impurities [14].

Using methods of measuring DC resistivity and microwave absorption, we studied the effect of spin fluctuations on the scattering of current carriers in the $BaFe_2As_2$ crystals with different cobalt concentrations. The method of growing crystals is described in detail in the article [15], and their transport and magnetic properties are presented in Refs. [15, 16].

3. Results and discussion

We will use below the temperature dependence of DC resistivity and microwave absorption, obtained for two Ba(Fe_{1-x}Co_x)₂As₂ crystals with x = 0.05 (underdoped) and x = 0.075 (over-doped), to demonstrate their salient features and analysis.

The resistivity versus temperature for the $Ba(Fe_{0.95}Co_{0.05})_2As_2$ sample is shown in Figure 1. This dependence has all the features characteristic of crystals $Ba(Fe_{1-x}Co_x)_2As_2$ with a cobalt impurity concentration less than optimal, x < 0.07 [1–4, 17]. Namely: metallic run with a positive slope in the interval $T_s < T < 300 \,\mathrm{K}$ with linear dependence $\rho(T) \propto T$ at $T > 115 \,\mathrm{K}$ and deviation from linearity when approaching the point of structural transition $T_s \approx 75 \,\mathrm{K}$ upon decreasing temperature. When T_s is reached, the resistance makes a sharp jump upward, after which it changes slightly, then sharply drops to 0 upon transition to the superconducting state at $T = T_c$. It is generally accepted that the $\rho(T)$ dependence is described by the Equation (2) and its linear part is due to carrier scatter by spin fluctuations [1-3,7,17]. The presence of spin fluctuations and their effect on the transport and magnetic properties of iron arsenides has been confirmed by many experimental methods, including inelastic neutron scattering (INS) [18–20] and nuclear magnetic resonance (NMR) [16,17,21,22]. In particular, they determine the unusual form of the temperature dependence of magnetic susceptibility: $\chi_0(T) \propto T$ instead of the Curie-Weiss law. This form of dependence is due to that the exchange interaction J_1 between the magnetic moments of iron ions in the FeAs layers does not lead to long-range magnetic ordering even when the temperature drops below Θ ($k_{\rm B}\Theta \approx J_1 = 43 \,{\rm meV}$ [19]). It is because of the prohibition of such ordering in low-dimensional systems (the Mermin-Wagner theorem [23]),



Figure 1. Temperature dependence of the resistivity of the $Ba(Fe_{0.95}Co_{0.05})_2As_2$ crystal (diamonds). The dashed straight line is drawn along the experimental points obtained at high temperatures, T > 115 K. Red solid line is the fit of Equation 2 to data points of temperature range from 75 K to 300 K.

and the FeAs layers in Ba(Fe_{1-x}Co_x)₂As₂ are quasi-2D due to the weak connection J_{\perp} between layers. At the same time, sufficiently strong antiferromagnetic correlations lead to the formation of singlet pairs and a decrease in magnetic susceptibility with lowering temperature [24–26].

According to popular opinion, at low temperatures ($T < 100 \,\mathrm{K}$), the electron-electron scattering mechanism becomes more efficient than scattering by spin fluctuations, and the $\rho(T)$ dependence turns into quadratic [3,5,27]. The reason for this crossover is not entirely clear and leaves doubt as to its correctness. Here we propose another idea to explain the behavior of the $\rho(T)$ dependence at low temperatures.

In our opinion, it is more natural to associate the deviation from linearity with the influence of nematic fluctuations. The fact is that the magnetic fluctuations, which determine the resistivity value, have a highly anisotropic stripe-type order parameter. Therefore, the scattering rates on them for two crystallographic directions (a and b) are very different: $\rho^b > \rho^a$. A theoretical description of this effect is given in Ref. [8]. This is experimentally confirmed by measurements of the resistivity in two directions, ρ^a and ρ^b , of Ba(Fe_{1-x}Co_x)₂As₂ crystals subjected to uniaxial compression (tension), which leads to their detwinning [28, 29]. As a result, the dependences $\rho^a(T)$ and $\rho^b(T)$ are very different from each other both at $T < T_s$ and at $T > T_s$, and the temperature at which resistivity anisotropy appears is tens of degrees higher than T_N and T_s . In this case, $\rho^a(T)$ remains practically linear as the temperature decreases almost to the superconducting transition T_c , while $\rho^b(T)$ deviates from linearity with the emergence of nematic fluctuations and experiences an upturn at temperature close to T_N . In twinned crystals, which have not undergone uniaxial compression, $\rho(T)$ behaves as something between $\rho^a(T)$ and $\rho^b(T)$, that is, all deviations from linearity also occur. They are not as large as $\rho^b(T)$, but they are quite comparable to it in the degree of nematic fluctuation effect. Thus, the behavior of $\rho(T)$ can be well explained by the influence of nematic (anisotropic magnetic) fluctuations, without using the assumption of a transition to the Fermi-liquid regime.

As mentioned above, microwave absorption data provide additional information about fluctuations. The temperature dependence of the MWA amplitude for the $Ba(Fe_{0.95}Co_{0.05})_2As_2$

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sample is presented in Figure 2 along with $\rho(T)$. To analyze the $A_{\text{MWA}}(T)$ behavior, Equation (3) with definitions (4) and (5) is used. In so doing, we assume a linear dependence of $\rho(T)$ and $\chi_0(T)$. The temperature dependence of the scattering rate is used in the form proposed in Ref. [3]:

$$\tau_{\rm sf}^{-1}(T) = \frac{a \cdot T}{(T+\Theta)^{1/2}} + b \cdot T^2,\tag{6}$$

where $\Theta \approx J_1/k_{\rm B} \approx 500 \,\mathrm{K}$ is the mean-field temperature of magnetic ordering in the *ab* plane that did not occur due to the reasons noted above (value of J_1 is taken from Ref. [19]). The first term on the right side of the Equation (6) is related to scattering by spin fluctuations, and the second one describes electron-electron scattering. The result of fitting is shown in Figure 2 with a solid black curve. Note that when calculating without restricting the parameters, the value of the parameter *b* is always less than *a* by several orders of magnitude, and if we take b = 0, then the calculated curve virtually does not change. This indicates that there is no need to involve the Fermi-liquid scattering mechanism to describe ohmic losses and microwave absorption in iron pnictides, despite the fact that the $A_{\rm MWA}(T)$ dependence is nonlinear. The magnitude of $\tau_{\rm sf}^{-1}$ will be discussed below.



Figure 2. Temperature dependence of the MWA amplitude (red diamonds) and resistivity (blue curve) of the $Ba(Fe_{0.95}Co_{0.05})_2As_2$ crystal. Black solid line is the fit of Equation 3 to the MWA data points of temperature range from 70 K to 200 K.

The change of temperature dependence of resistivity and microwave absorption in a crystal Ba(Fe_{1-x}Co_x)₂As₂ when the cobalt concentration increases to values higher than optimal (x > 0.07) will be considered below using the example of the Ba(Fe_{0.925}Co_{0.075})₂As₂ sample. In accordance with the phase diagram [1,15], neither a structural transition nor magnetic ordering occurs in a sample of such composition. The dependences $\rho(T)$ and $A_{MWA}(T)$ obtained for this sample, presented in Figure 3, are well consistent with this statement. There are no features on both curves due to changes in structure or magnetic state. There is only a sharp drop in value (to 0 in the case of resistivity) when the sample transits to the superconducting state, $T_c = 24$ K. $\rho(T)$ has a fairly long linear section, $100 \div 300$ K, and upturn at T = 43 K. $A_{MWA}(T)$ is nonlinear over the entire measurement temperature range, from T_c up to 165 K. The calculation of this dependence using Equation (3) and the definitions of its parameters by formulas (4-6) is shown by black curve in the Figure 3.



Figure 3. Temperature dependence of resistivity (blue solid line) and the MWA amplitude (red circles) of the Ba(Fe_{0.925}Co_{0.075})₂As₂ crystal. Black solid line is the fit of Equation 3 to the MWA data in the temperature range from 60 K to 165 K. A dashed straight line is drawn to show a linear section of $\rho(T)$.

One of the most important parameters of the studied compounds, which determines the value and behavior of both resistivity (Eq. 1) and microwave absorption (Eq. 3), is the scattering rate τ^{-1} (or scattering time τ). The parameter values obtained by fitting the theoretical dependence $A_{\text{MWA}}(T)$ (Eq. (3)) to the experimental data were as follows: τ_{sf}^{-1} changes from $2.1 \cdot 10^{10} \,\mathrm{s}^{-1}$ to $6.3 \cdot 10^{10} \,\mathrm{s}^{-1}$ with increasing temperature in the range of $75 - 185 \,\mathrm{K}$ in case of sample Ba(Fe_{0.95}Co_{0.05})₂As₂. And it varies from $2 \cdot 10^{10} \,\mathrm{s}^{-1}$ to $1.5 \cdot 10^{11} \,\mathrm{s}^{-1}$ at $T = 60 \div 185 \,\mathrm{K}$ for the $Ba(Fe_{0.925}Co_{0.075})_2As_2$ sample. The obtained values of scattering rates are significantly lower than their estimates made for similar samples based on DC transport measurements and presented in literature: $3 \cdot 10^{13} \text{ s}^{-1}$ at 300 K [27] and $3.5 \cdot 10^{12} \text{ s}^{-1}$ at 100 K [30]. This discrepancy can be understood if we take into account that estimates from transport measurement data give the total dissipation rate τ^{-1} , which is determined by all mechanisms, including an impurity one. And the values obtained from the MWA fitting relate only to scattering by spin fluctuations, $\tau_{\rm sf}^{-1}$. This explanation is consistent with the results of ultrafast time-resolved polarimetry [31], which revealed a two-step recovery of spin system of $Ba(Fe_{1-x}Co_x)_2As_2$ excited by a femtosecond pulse of polarized light. At the first step, rapid thermalization of charges occurs within 10^{-13} s after the exciting pulse. The second step is a slow recovery via nematic fluctuations over a period of $10^{-11} - 10^{-10}$ s. It can be seen that the $\tau_{\rm sf}^{-1}$ values obtained from the $A_{\rm MWA}(T)$ analysis correspond to the second stage of relaxation of the spin system.

4. Conclusion

In order to clarify what mechanisms of current carrier scattering determine the magnitude and behavior of resistance and microwave absorption in crystals $\operatorname{Ba}(\operatorname{Fe}_{1-x}\operatorname{Co}_x)_2\operatorname{As}_2$, we analyzed the temperature dependences $\rho(T)$ and $A_{\mathrm{MWA}}(T)$ of samples with cobalt concentration x = 0.05 and 0.075. The results of analysis allow us to conclude that the deviation of the $\rho(T)$ dependence from the linear behavior at $T \leq 100 \,\mathrm{K}$ is not related to the transition to the Fermi-liquid regime with $\rho(T) \propto T^2$, but is due to the emerging nematic fluctuations. The rate of scattering by spin fluctuations τ_{sf}^{-1} , determined by describing the temperature dependence of the MWA amplitude, indicates that they take nematic character as the temperature decreases down to the SDW ordering point.

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