



# Surface impedance in the antiferromagnetic and superconducting states of underdoped BaFe<sub>1.93</sub>Ni<sub>0.07</sub>As<sub>2</sub> crystals



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## ABSTRACT

Measurements of the real  $R$  and imaginary  $X$  parts of the surface impedance were performed in underdoped BaFe<sub>1.93</sub>Ni<sub>0.07</sub>As<sub>2</sub> crystals in the frequency range 10 MHz–1.5 GHz. The establishment of the antiferromagnetic order at  $T_N \sim 50$  K gives rise to anomalous increase of electron scattering time. Drude type conductivity yields  $X$  and  $R$  differ from each other. The increase of the real conductivity  $\sigma_1$  in the superconducting state is attributed to a rapid decrease of the quasiparticle scattering time. This result gives evidence of coexistence of superconductivity and antiferromagnetism.

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## 1. Introduction

The underdoped BaFe<sub>1.93</sub>Ni<sub>0.07</sub>As<sub>2</sub> exhibits a structural transition at  $T_S \sim 70$  K, an antiferromagnetic transition at  $T_N \sim 50$  K and a superconducting transition at  $T_C \sim 16$  K [1–4]. NMR measurements give clear evidence of coexistence of superconductivity and antiferromagnetism [1]. The spin-lattice relaxation shows a critical slowing down around  $T_N$  and a further strong decrease around the superconducting transition. A number of surface impedance measurements have been published on iron based superconductors. They give information on London penetration depth and quasiparticle scattering [5–9]. The power temperature dependence of the London penetration depth in the  $ab$  plane  $\lambda(T) - \lambda(0) \sim T^n$  with  $n \sim 2$ –2.8 is attributed to pair breaking scattering in the so-called  $s^\pm$  superconductivity [5–8]. The sharp increase of the real part of the conductivity below  $T_C$  was explained by a rapid decrease of the quasiparticle scattering rate for the optimally Co and Ni doped crystals [7–9].

Here we report surface impedance measurements on underdoped BaFe<sub>1.93</sub>Ni<sub>0.07</sub>As<sub>2</sub> in the frequency range 10 MHz–1.5 GHz. Careful analysis of the impedance measurements permits us to

observe that the real  $R$  and imaginary  $X$  parts of the surface impedance of underdoped BaFe<sub>1.93</sub>Ni<sub>0.07</sub>As<sub>2</sub> differ from each other in the antiferromagnetic state. A large decrease of the scattering rate  $\tau^{-1}$  of the charge carriers produced by the establishment of the antiferromagnetic order gives a complex Drude conductivity  $\sigma = \sigma_0 / (1 + j\omega\tau)$  where  $\omega$  is the measuring angular frequency with  $\omega\tau \sim 1$  [10]. Complex conductivity induces  $R \neq X$ .

## 2. Experiment

The crystals were grown using a Fe/Ni–As self flux method, details are given in [4]. The samples are platelike with the plates perpendicular to the crystallographic axis  $c$ . The same surface impedance measurement technique was used for optimally and over doped BaFe<sub>2-x</sub>Ni<sub>x</sub>As<sub>2</sub> crystals in reference [9]. The samples are placed inside a copper coil situated at the end of a coaxial line. The radio frequency magnetic field is applied parallel to the  $ab$  plane. Non resonant measurements of the resistance and inductance of the coil were performed with an automated impedance analyzer Agilent 4395 in the frequency range 10–100 MHz. Resonant measurements of the LC resonant frequency,  $L = 0.2 \mu\text{H}$ ,  $C = 50 \text{ fF}$  quality factor 80, were performed at 1.5 GHz with a Hewlett Packard 8720B network analyzer. Resistance and inductance were measured separately in the absence of the sample and were subtracted from measurements with the sample present.

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The inductance of the coil is given by [11]

$$Z = R_0 + \frac{k^2 L_0 L_2 \omega^2 R}{|R + j(X + L_2 \omega)|^2} + j \left[ L_0 - \frac{k^2 L_0 L_2 \omega (X + L_2 \omega)}{|R + j(X + L_2 \omega)|^2} \right] \omega \quad (1)$$

where  $R$  and  $X$  are the real and imaginary parts of the surface impedance of the sample,  $R_0$  and  $L_0 \sim 0.2 \mu\text{H}$ , are the resistance and inductance of the empty coil and  $\omega$  is the angular frequency. Coupling  $k$  and geometrical  $L_2$  factors were evaluated from measurements at high temperatures  $\sim 100 \text{ K}$ ,  $k^2 \sim 0.1$  and  $L_2 \sim 0.6 \text{ nH}$ .  $R$  and  $X$  are deduced from Eq. (1). At high temperatures

$$Z = \sqrt{\frac{j\mu_0 \omega}{\sigma_{\text{HT}}}} \quad \text{with} \quad X_{\text{HT}} = R_{\text{HT}} = \sqrt{\mu_0 \rho_{\text{dc}} \omega / 2} \quad (2)$$

where  $\mu_0$  is the magnetic permeability of vacuum and  $\sigma_{\text{HT}} = 1/\rho_{\text{dc}}$  is the conductivity at  $\sim 100 \text{ K}$ ,  $\rho_{\text{dc}} \sim 0.2 \text{ m}\Omega \text{ cm}$  is the measured dc resistivity [4]. At  $100 \text{ K}$ , a skin depth  $\delta = \sqrt{2/\mu_0 \sigma_{\text{HT}} \omega} \sim 50 \mu\text{m}$  is estimated at  $100 \text{ MHz}$ .

### 3. Results and discussion

The temperature dependence of  $R$  and  $X$  normalized to the value  $R_{\text{HT}}$  measured at  $100 \text{ K}$  are shown in Figs. 1 and 2.  $X$  and  $R$  differ from each other in the superconducting state below  $T_c$  and in the antiferromagnetic state below the Neel transition  $T_N \sim 50 \text{ K}$ . Meanwhile  $X=R$  is found in the high temperature range above  $T_N$ .

#### 3.1. Antiferromagnetic state in the temperature range $T_c < T < T_N$

$X$  and  $R$  differ from each other in the antiferromagnetic state. An increase of the electron scattering time  $\tau_m$  in the antiferromagnetic state originates a complex Drude type conductivity  $\sigma = \sigma_0 / (1 + j\omega\tau_m)$  [10], where  $\sigma_0$  is the dc conductivity

Surface impedance is now given by

$$Z = R + jX = \sqrt{\frac{j\mu_0(1 + \omega\tau_m)\omega}{\sigma_0}} \quad (3)$$

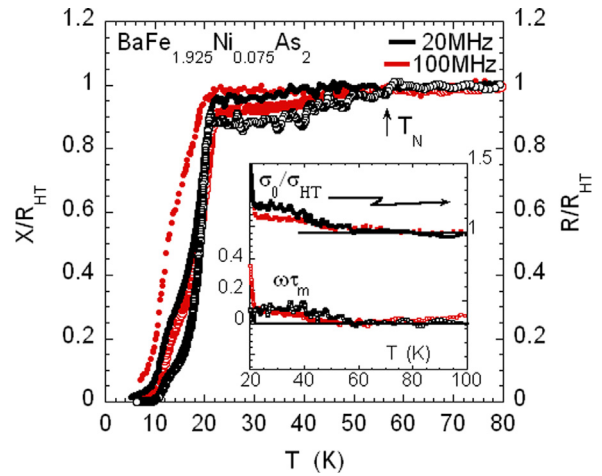
The parameters  $\omega\tau_m$  and  $\sigma_0/\sigma_{\text{HT}}$  are deduced from the measurements of  $X/R_{\text{HT}}$  and  $R/R_{\text{HT}}$  using the following relations deduced

from Eq. (3)

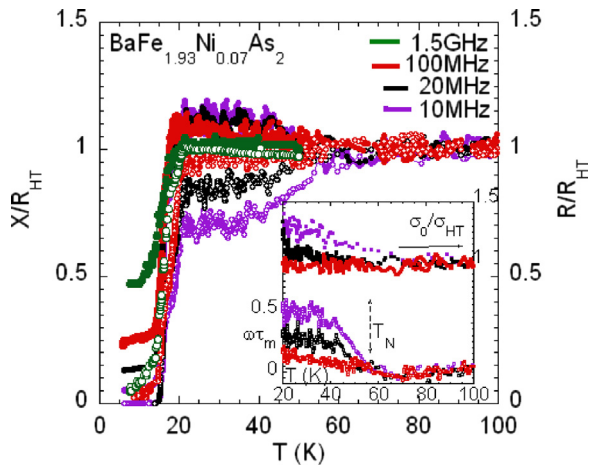
$$\frac{\sigma_0}{\sigma_{\text{HT}}} = \frac{R_{\text{HT}} R_{\text{HT}}}{R X} \quad \text{and} \quad \omega\tau_m = \frac{(X/R_{\text{HT}})^2 - (R/R_{\text{HT}})^2}{2(R/R_{\text{HT}})(X/R_{\text{HT}})} \quad (4)$$

The resulting  $\omega\tau_m$  and  $\sigma_0/\sigma_{\text{HT}}$  parameters are shown in Figs. 1 and 2. For  $T > T_N$ ,  $\omega\tau_m = 0$  is consistent with  $X_{\text{HT}} = R_{\text{HT}}$  in the high temperature state. In the antiferromagnetic state  $\tau_m$  increases with decreasing temperature from  $T_N$  to  $20 \text{ K}$ . A saturating value of  $\omega\tau_m \sim 0.12, 0.26$  and  $0.5$  is found around  $20 \text{ K}$  at  $10, 20$  and  $100 \text{ MHz}$ . Within our experimental resolution no difference between  $R$  and  $X$  is detected at  $1.5 \text{ GHz}$  in the temperature range above  $T_c$ . For Ni doping level  $x=0.075$   $\omega\tau_m \sim 0.1$  is obtained at  $20$  and  $100 \text{ MHz}$  (Fig. 2).

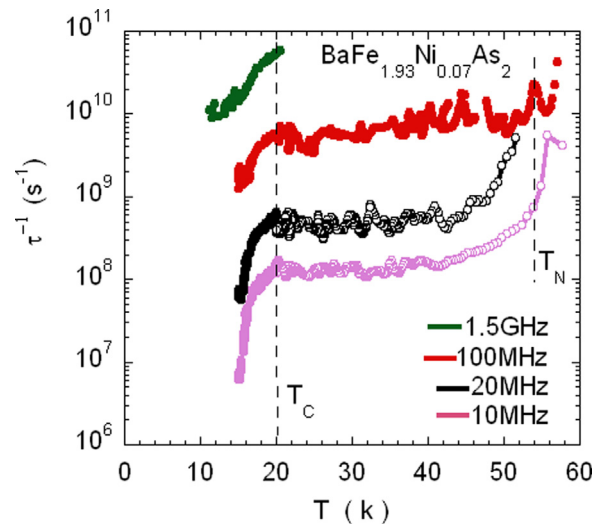
The temperature dependence of the relaxation rate  $\tau_m^{-1}$  deduced from Fig. 1 is shown in Fig. 3. Increase of  $\tau_m^{-1}$  with



**Fig. 2.** Temperature variation of  $X/R_{\text{HT}}$  (filled symbols) and  $R/R_{\text{HT}}$  (open symbols) of surface impedance,  $R_{\text{HT}}$  is obtained at  $100 \text{ K}$  for the underdoped  $\text{BaFe}_{1.925}\text{Ni}_{0.075}\text{As}_2$  crystals at  $20 \text{ MHz}$  (black),  $100 \text{ MHz}$  (red). Inset: Temperature dependence of  $\sigma_0/\sigma_{\text{HT}}$  normalized to the value  $\sigma_{\text{HT}}$  at  $100 \text{ K}$  and of  $\omega\tau_m$  with  $\omega = 2\pi F$ ,  $F = 20$  and  $100 \text{ MHz}$ . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)



**Fig. 1.** Temperature variation of  $X/R_{\text{HT}}$  (filled symbols) and  $R/R_{\text{HT}}$  (open symbols) of surface impedance,  $R_{\text{HT}}$  is obtained at  $100 \text{ K}$  for the underdoped  $\text{BaFe}_{1.93}\text{Ni}_{0.07}\text{As}_2$  crystals at  $10 \text{ MHz}$  (violet),  $20 \text{ MHz}$  (black),  $100 \text{ MHz}$  (red),  $1.5 \text{ GHz}$  (green). Inset: Temperature dependence of  $\sigma_0/\sigma_{\text{HT}}$  normalized to the value  $\sigma_{\text{HT}}$  at  $100 \text{ K}$  and of  $\omega\tau_m$  with  $\omega = 2\pi F$ ,  $F = 10, 20$  and  $100 \text{ MHz}$ . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)



**Fig. 3.** Temperature dependence of the electron scattering rate  $\tau_m^{-1}$  in the temperature range  $20 \text{ K} < T < T_N$  and of the quasiparticle scattering rate  $\tau^{-1}$  deduced from the conductivity  $\sigma_1$  in the superconducting state for the underdoped  $\text{BaFe}_{1.93}\text{Ni}_{0.07}\text{As}_2$ . At each frequency we take  $\tau^{-1}(20 \text{ K}) = \tau_m^{-1}(20 \text{ K})$ .

increasing frequency or decreasing skin depth  $\delta$  is found. Such an effect could be attributed to an additional scattering of electrons by surface defects whose contribution increases with the decreasing skin depth. Conductivity  $\sigma_0$  increases slightly with the decreasing temperature from  $T_N$  to 20 K Figs. 1 and 2.

### 3.2. Superconducting state

Our resistance measurements are well resolved in the vicinity of the superconducting transition.

But below 15 K our experimental resolution is not sufficient to extract the intrinsic resistance.

Surface impedance  $Z$  is related to the real  $\sigma_1$  and imaginary  $\sigma_2$  parts of the conductivity:

$$Z = R + jX = \sqrt{\frac{j\mu_0\omega}{\sigma_1 - j\sigma_2}} \quad (5)$$

$\sigma_1$  and  $\sigma_2$  are normalized to the value  $\sigma_1(20\text{ K}) = \sigma_0(20\text{ K}) \sim \sigma_{HT}$  at 20 K.

The real  $\sigma_1$  and imaginary  $\sigma_2$  parts are given by inverting Eq. (5).

$$\frac{\sigma_1}{\sigma_{HT}} = \frac{4RX/R_{HT}^2}{[(R/R_{HT})^2 + (X/R_{HT})^2]^2} \quad \frac{\sigma_2}{\sigma_{HT}} = 2 \frac{(X/R_{HT})^2 - (R/R_{HT})^2}{[(R/R_{HT})^2 + (X/R_{HT})^2]^2} \quad (6)$$

Temperature dependences of  $\sigma_1/\sigma_{HT}$  and  $\sigma_2/\sigma_2(0)$ , with  $\sigma_2(0) = \sigma_2(T=0)$  are shown in Fig. 4.

The conductivity  $\sigma_2/\sigma_2(0)$  is related to the London penetration depth [4–9] and follows a  $T^2$  temperature dependence:

$$\frac{\sigma_2}{\sigma_2(0)} = \frac{\lambda(0)^2}{\lambda(T)^2} \quad \text{and} \quad \frac{\sigma_2}{\sigma_2(0)} \approx 1 - \left(\frac{T}{T_C}\right)^2 \quad (7)$$

The conductivity  $\sigma_1$  increases with decreasing temperature below  $T_C \sim 16$  K to a maximum of about 2–10 times its normal value and this maximum decrease with increasing frequency from 10 MHz to 1.5 GHz. The strong rise in the conductivity  $\sigma_1$  below  $T_C$  results from a temperature dependent scattering rate decreasing rapidly with temperature [7–9]. A smaller broad variation is observed between 16 K and 20 K. The phenomenological two fluid model is used to extract the quasiparticle scattering time  $\tau$  [12,13]. The superconducting density,  $n_S$  is related to the London penetration depth  $n_S = \lambda(0)^2/\lambda_2$ . The normal density  $n_N$  is given by  $n_N = 1 - n_S$ , the total carrier density being 1. It results that the temperature

dependence of  $n_N$  in the underdoped  $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$  crystals follows the  $(T/T_C)^2$  temperature dependence.  $\sigma_1$  is proportional to the product  $n_N\tau$  [12] which gives,  $\sigma_1 \sim (T/T_C)2\tau$ . Consequently scattering rate  $\tau^{-1}$  is given by

$$\frac{\tau^{-1}(T)}{\tau^{-1}(T_C)^{-1}} = \left(\frac{T}{T_C}\right)^2 \frac{\sigma_1^{-1}(T)}{\sigma_1^{-1}(T_C)} \quad (8)$$

where  $\tau^{-1}(T_C)$  is the scattering rate at  $T_C \sim 16$  K.

In the temperature range 16–20 K (Fig. 4)  $\sigma_2 \sim 0$  gives  $n_N \sim 1$  and Eq. (8) is reduced to

$$\frac{\tau^{-1}(T)}{\tau^{-1}(20\text{ K})} = \frac{\sigma_1^{-1}(T)}{\sigma_1^{-1}(20\text{ K})} \quad (9)$$

At 20 K the scattering rate  $\tau^{-1}(20\text{ K})$  is equal to  $\tau_m^{-1}(20\text{ K})$  which is the relaxation rate determined with Eq. (4). The scattering rates deduced at the different frequencies are shown in Fig. 3. At 1.5 GHz,  $\tau^{-1}(20\text{ K}) \sim 6 \times 10^{10}$  was arbitrary taken.

Finally significant decrease of the electron scattering rate is observed around  $T_N$  (Fig. 3) when magnetic order sets in. Below  $T_C$ , strong decrease of the quasiparticle scattering rate is found in the superconducting state. This result is similar to the temperature dependence of  $1/T_1$  [1]. Evidence for coexistence of superconductivity and antiferromagnetism is given.

The superconducting transition is quite broadened in the temperature range 16–20 K for the underdoped crystals in contrast with the optimally and over doped crystals [9]. This result is in agreement with the broad elastic anomaly of the C33 mode observed with the same crystals [14]. The purely electronic effects are strongly affected by structural instabilities near the superconducting transition in the Ni and Co underdoped  $\text{BaFeAs}_2$  crystals [15].

## 4. Conclusion

Establishment of antiferromagnetic order in underdoped  $\text{BaFe}_{1.93}\text{Ni}_{0.07}\text{As}_2$  crystals gives rise to an anomalous increase of electron scattering time which leads that the real and imaginary parts of complex surface impedance differ from each other  $X > R$ . The increase of the real conductivity  $\sigma_1$  in the superconducting state is attributed to a strong decrease of the quasiparticle scattering time. This result gives evidence for coexistence of superconductivity and antiferromagnetism.

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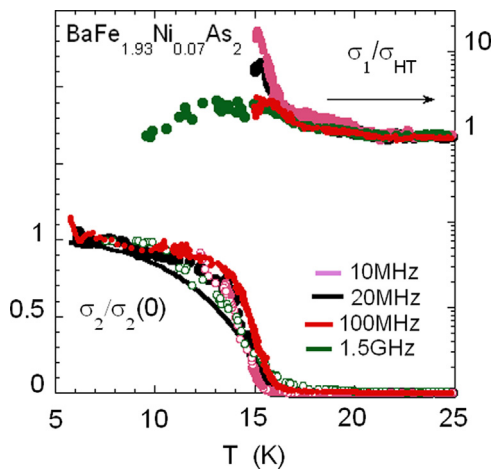


Fig. 4. Temperature dependence of the real  $\sigma_1/\sigma_{HT}$  (filled symbols) and imaginary  $\sigma_2/\sigma_{HT}$  (open symbols) parts of the effective conductivity,  $\sigma_{HT}$  is the conductivity at 100 K, for underdoped  $\text{BaFe}_{1.93}\text{Ni}_{0.07}\text{As}_2$  at 10 MHz, 20 MHz, 100 MHz and 1.5 GHz. Solid line  $\sigma_2/\sigma_2(0)$  is calculated with  $1 - (T/16)^2$ .

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