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Direct observation of the superconducting energy gap in the optical conductivity of the iron pnictide superconductor $Ba(Fe_{0.9}Co_{0.1})_2As_2$

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The temperature-dependent optical reflectivity and complex transmissivity of an epitaxially grown $Ba(Fe_{0.9}Co_{0.1})_2As_2$ thin film were measured over a wide frequency range (4–35 000 cm^{-1}). The opening of the superconducting gap $2\Delta_0=(3.7\pm0.3)$ meV is *directly* observed by vanishing optical conductivity at 30 cm^{-1} for $T<T_c=20$ K. While in this range the measured temperature- and frequency-dependent electrodynamic properties agree well with the BCS predictions of a nodeless order parameter, unexpectedly a strong quasiparticle absorption shows up below 1.5 meV. The spectral weight of the condensate 1.94×10^7 cm^{-2} corresponds to a penetration depth $\lambda=3600$ Å.

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I. INTRODUCTION

Soon after the discovery of superconductivity in iron pnictides,[1] the epitaxial growth of LaFeAsO films was reported[2,3] and superconductivity observed in thin films of Co-doped $SrFe_2As_2$.[4,5] By now the homogeneity of the films and the upper critical field have increased to make iron pnictides interesting for technological applications.[6–8] In particular cobalt-doped $BaFe_2As_2$ seems to be suitable for producing high-quality thin films which are stable in air,[8] can be template engineered,[9] or tuned in T_c by epitaxial growths of strained films.[10]

Besides potential applications, thin films are advantageous for investigations of fundamental problems due to their large area, in particular if single crystals of sufficient quality, homogeneity, and size are limited. As far as optical experiments are concerned, only thin films give the opportunity to perform transmission measurements and in this way be much more sensitive to probe the electrodynamic properties of the normal and superconducting states.[11,12] Issues such as the spectral weight distribution, the universal conductivity background, and in particular on the superconducting gaps, on states in the gap, nodes in the order parameter and quasiparticle relaxation are addressed by our optical experiments on Ba-$(Fe_{0.9}Co_{0.1})_2As_2$ thin films.

II. EXPERIMENTAL DETAILS AND RESULTS

$Ba(Fe_{1-x}Co_x)_2As_2$ films were deposited on a (001)-oriented $(La,Sr)(Al,Ta)O_3$ substrate by pulsed laser deposition, where the $Ba(Fe_{0.9}Co_{0.1})_2As_2$ target was ablated with 248 nm KrF radiation under UHV conditions.[10] The films grow with a very smooth surface of an rms roughness better than 12 nm, as measured by atomic force microscopy (AFM). The film thickness was monitored *in situ* by a quartz balance, and finally measured by AFM and ellipsometry to be $d=90$ nm. The phase purity was checked by x-ray and EDS. Standard four-probe method was utilized to measure the dc resistivity and determine the superconducting transition: from the onset at 22 K with a transition width of 2 K, we have chosen $T_c=20$ K (inset of Fig. 1). The absolute value and temperature dependence of $\rho(T)$ is very similar to results obtained on single crystals.[13]

Using different optical methods, we performed experiments in the frequency range from 4 to 35 000 cm^{-1} and at various temperatures down to 5 K. In the THz range (4 to 40 cm^{-1}) the complex transmissivity (transmission amplitude and phase) was measured utilizing a Mach-Zehnder arrangement, as depicted in Fig. 2(b).[14] Between 20 and 15 000 cm^{-1} the reflectivity was investigated by Fourier transform infrared spectroscopy; a gold mirror served as reference. The spectra were extended up to the ultraviolet by room-temperature ellipsometric data (6000–35 000 cm^{-1}). In order to determine the properties of the film, we measured the optical parameters of a bare $(La,Sr)(Al,Ta)O_3$ substrate over the entire frequency and temperature ranges.

In Fig. 1(a) the optical reflectivity is plotted in a wide frequency range for selected temperatures. In particular in the far-infrared range the phonons of the substrate become obvious. The small film thickness and moderate conductivity prevent the use of a Kramers-Kronig procedure for obtaining the conductivity and permittivity spectra of the semioblique film. For the further analysis we therefore employ a two-layer model that consists of the $(La,Sr)(Al,Ta)O_3$ substrate with thickness of 1.023 mm and optical parameters determined beforehand, covered by the thin film of $Ba(Fe_{0.9}Co_{0.1})_2As_2$. Using Fresnel's equations[11] we can analyze the intrinsic optical properties of the film. In the THz range (4–40 cm^{-1}) where data for the transmission and phase shift are available, a corresponding analysis was performed that allowed us to directly determine the values of dielectric permittivity and conductivity, with the experimental uncertainties strongly dependent on the values of $\epsilon(\omega,T)$ and $\sigma(\omega,T)$.[14] The same model was used to evaluate the optical response of the film at higher frequencies. Eventually a self-consistent fit of the conductivity and permittivity spectra at THz frequencies and the reflectivity spectra at higher frequencies yields the overall behavior of the conductivity as presented in Fig. 1(b).

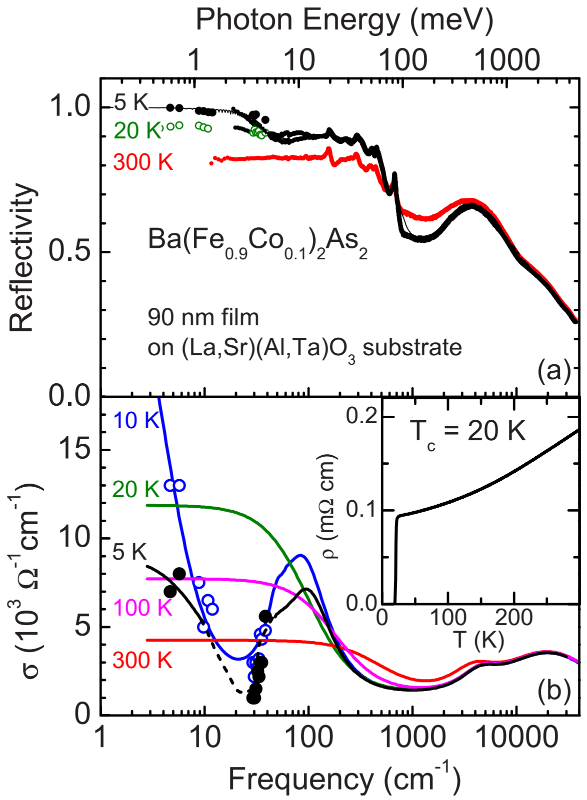

FIG. 1. (Color online) (a) Reflectivity of a 90 nm $Ba(Fe_{0.9}\text{-}Co_{0.1})_2As_2$ film on a 1 mm (La,Sr)(Al,Ta)O_3 substrate measured in a wide frequency range at various temperatures. The dots between 4 and 40 cm^{-1} are calculated from the transmission measurements. The thin gray line indicates the fit of the 5 K spectrum by the Drude-Lorentz analysis. (b) Optical conductivity of $Ba(Fe_{0.9}Co_{0.1})_2As_2$ obtained from the Drude-Lorentz analysis of the reflection and transmission spectra. The dots are directly calculated from transmission and phase measuring by the Mach-Zehnder interferometer. The dashed part of the 5 K curve between 10 and 50 cm^{-1} indicates that a simple Lorentz shape does not mimic the superconducting gap properly since $\sigma(\omega)$ basically vanishes abruptly at 30 cm^{-1}. Below 10 cm^{-1} a strong quasiparticle absorption is observed which seem to disappear only well below 5 K. The inset shows the dc resistivity $\rho(T)$ of our sample with T_c=20 K.

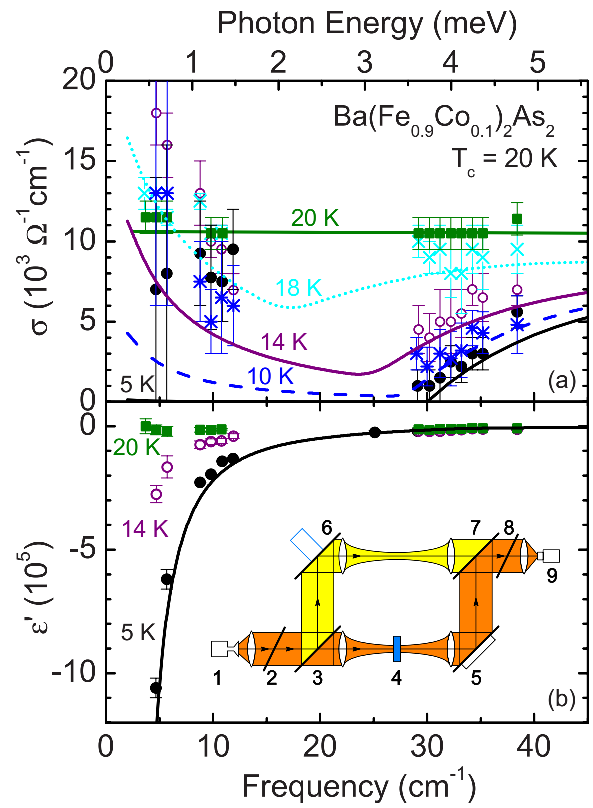

FIG. 2. (Color online) Optical properties of $Ba(Fe_{0.9}\text{-}Co_{0.1})_2As_2$ in the THz range where transmission and phase measurements directly yield the (a) conductivity and (b) permittivity spectra. The lines in panel (a) are calculated from the BCS theory assuming a complete opening of the gap at 30 cm^{-1} over the entire Fermi surface. A sketch of the Mach-Zehnder arrangement is included in panel (b); the reference arm (yellow, light gray) is additionally utilized to determine the phase shift. 1, radiation source; 2,8, wire-grid polarizer; 3,7, beam splitter; 4, sample; 5, fixed mirror; 6, movable mirror; and 9, detector.

The properties of the $Ba(Fe_{0.9}Co_{0.1})_2As_2$ film in the normal state are described by two Drude terms, a narrow σ_N and a broad one σ_B, corresponding to two types of charge carriers as suggested in Ref. 15. In addition, two Lorentz terms account for the interband transitions above 2000–3000 cm^{-1}. In the superconducting state, two additional Drude terms are introduced: one with a tiny scattering rate to model the δ-function (Cooper pair response) obvious in the permittivity spectrum and another term to describe the quasiparticle contribution to the below-gap conductivity. The optical response at energies around the superconducting gap is mimicked with two Lorentzians since an appropriate model-independent expression is missing in fit programs. The resulting curves $\sigma(\omega,T)$ are plotted in Fig. 1(b) for selected temperatures.

The most important finding of our investigation is the distinct opening of the superconducting gap which is directly seen in the drop of $\sigma(\omega,T)$ around 30 cm^{-1} upon cooling below T_c, clearly depicted in Fig. 1(b) and in more detail in Fig. 2(a) where we also indicated the uncertainty of our data; for T=5 K the conductivity has completely vanished at $\omega/(2\pi c)\approx 30$ cm^{-1}. From this figure the value of the superconducting energy gap can be determined as $2\Delta_0$ =3.7 meV ± 10%. Due to remaining quasiparticles, the conductivity is large below 10 cm^{-1} and even exceeds the normal-state value for $T>T_c/2$. In spite of the large experimental uncertainties, it seems to vanish gradually for $T\to 0$ but much slower than expected. The enormous drop of the low-frequency permittivity $\epsilon'(\omega,T)$ plotted in Fig. 2(b) evidences the inductive response of the superconducting condensate. It should be noted that the overall $\sigma(\omega,T)$-behavior of the $Ba(Fe_{0.9}Co_{0.1})_2As_2$ film is identical to findings on single crystals[13,15,16] and thus resembles the intrinsic and general optical behavior of 122 iron pnictides.

III. ANALYSIS AND DISCUSSION

In the metallic state ($T>20$ K) the optical conductivity of $Ba(Fe_{0.9}Co_{0.1})_2As_2$ has three major components: (i) the infrared peaks at 4400 and 20 800 cm^{-1} indicate interband transitions. (ii) The background $\sigma_B \approx 1300(\Omega\ cm)^{-1}$, which is best seen in the conductivity minimum around 1000 cm^{-1}, is more or less temperature independent; we model it with a Drude term, albeit its roll-off (the scattering rate and hence the plasma frequency) cannot be accurately determined.[17] (iii) Most important is the narrow Drude component $\sigma_N(\omega)$; the corresponding static conductivity grows and the peak becomes sharper as T decreases. Its spectral weight decreases by 15% when the temperature is reduced from 300 to 20 K.[18] We ascribe this to a slight enhancement of the effective mass due to electronic correlations as previously observed.[13,19]

For $T < T_c$ the THz conductivity dramatically decreases below 100 cm^{-1} due to the opening of the superconducting gap at 30 cm^{-1}, as seen in Fig. 1(b). The directly measured conductivity is displayed in more detail in Fig. 2(a) together with calculations of the BCS-based Mattis-Bardeen model.[11,20,21] The best description is obtained for $2\Delta_0 = 3.7$ meV, corresponding to $2\Delta_0/k_B T_c \approx 2.1$; this value is considerably lower than expected from mean-field theory, but similar to what has been determined from reflection measurements of single crystals.[15,16] In this region the shape of $\sigma(\omega)$ agrees well with the BCS prediction for a simple s-wave superconductor with no nodes in the order parameter. While an incomplete or anisotropic d-wave gap with nodes better fits the absorption below 10 cm^{-1}, it would cause a much larger contribution to $\sigma(\omega,T)$ in the gap-region around 30 cm^{-1} where the uncertainty of our data is quite small. Previous optical investigations[22,23] on hole-doped 122 iron pnictides draw similar conclusions. While our film of $Ba(Fe_{0.9}Co_{0.1})_2As_2$ is in the dirty limit, the K-doped compounds look more like a clean-limit superconductor, where the structure in $\sigma(\omega)$ is not directly the gap feature but influenced by bosonic excitations.

Below 10 cm^{-1} a very narrow peak builds up for $T < T_c$ due to the quasiparticle contribution to the conductivity; its intensity first increases and then diminishes as the quasiparticle number vanishes when $T \to 0$. It is not clear by now why this low-frequency absorption is much stronger than expected from theory for an isotropic and complete gap. In Fig. 3(a) the temperature dependence of $\sigma(\omega,T)$ is plotted for selected frequencies. From room temperature down to $T = 30$ K we find the THz conductivity basically not dependent on frequency, as expected for a normal metal; maybe small indications of fluctuations below 50 K can be identified. In the superconducting state, the conductivity at very low frequency $[\omega/(2\pi c) = 4.6\ cm^{-1}]$ increases strongly right below T_c, it passes through a maximum σ_{max} around 14 K $\approx 0.7\ T_c$, and then drops rapidly. As the frequency increases, this peak vanishes and only a simple drop of the conductivity is observed for $T < T_c$. In a very recent paper, Schachinger *et al.* discussed exactly this behavior and compared it with different models and parameters.[24]

The BCS theory predicts a so-called coherence peak in the electrodynamic absorption of superconductors;[11,21] it is considered as a hallmark of singlet superconductivity and most pronounced in the dirty limit and for s-wave symmetry of the order parameter. In contrast to theory and observations on conventional superconductors,[25,26] in the present case the quasiparticle peak is larger in amplitude and extends to lower temperatures, as already seen in Fig. 2(a). An even larger peak has been reported[27] from microwave experiments on K-doped $BaFe_2As_2$ and extensively discussed in Ref. 24. It is desired to conduct further experiments on different compositions and films in order to obtain more information on scattering mechanisms, the influence of multiple bands and cross scattering between the bands.

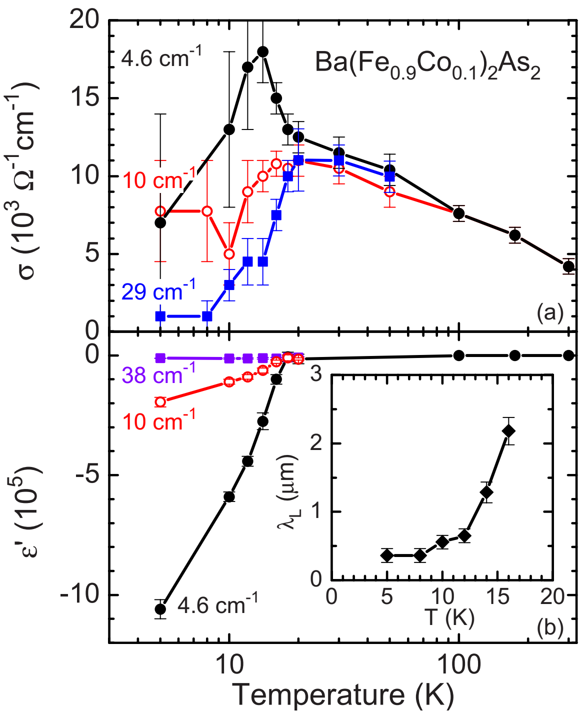

FIG. 3. (Color online) Temperature dependence of the low frequency (a) optical conductivity $\sigma(\omega,T)$, (b) permittivity $\epsilon(\omega,T)$, and penetration depth $\lambda(T)$. For the lowest frequencies $\sigma(T)$ clearly exhibits a maximum below T_c.

According to the Ferrell-Glover-Tinkham sum rule[11,21] the missing area

$$A = \int [\sigma^{(n)}(\omega) - \sigma^{(s)}(\omega)]\mathrm{d}\omega \quad (1)$$

between the conductivity in the normal and the superconducting state is collected in the δ peak of the condensate at $\omega = 0$. It is a measure of the penetration depth and gives $\lambda = c/\sqrt{8A} = (2500 \pm 700)$ Å. Alternatively the contribution of the superconducting carriers is probed by the permittivity ϵ' which for low frequencies goes as $1 - \epsilon' \propto (\omega_{ps}/\omega)^2$ in excellent agreement with the experimental results plotted in Fig. 2(b). Its development with temperature is displayed in the inset of Fig. 3(b). For $T \to 0$ we obtain $\lambda = (3600 \pm 500)$ Å in good agreement with the values we obtained for single crystals.[15,16] The spectral weight $(\omega_{ps}/2\pi c)^2 = (1.94 \pm 0.1) \times 10^7\ cm^{-2}$ of the superconducting condensate is in excellent agreement with the scaling relation suggested for cuprates.[28–30] We note that taking into account the below-gap contribution to the conductivity due to quasiparticles allows us to obtain more realistic values of the superconducting density compared to reflectivity experiments.

IV. CONCLUSIONS

Comprehensive optical investigation of $Ba(Fe_{0.9}$-$Co_{0.1})_2As_2$ thin films in a wide frequency and temperature range elucidate the electrodynamic properties in the superconducting state. The opening of the superconducting gap of $2\Delta_0 = 3.7$ meV; i.e., $2\Delta_0/k_BT_c = 2.1 \pm 10\%$ is *directly* observed by our THz transmission and phase measurements. In this region the temperature and frequency dependence of the conductivity is well described by the BCS theory assuming a complete isotropic gap. However, there remains a strong quasiparticle absorption below 10 cm^{-1}, which is not predicted by the simple model.

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[17] For our fit we used $1/(2\pi c\tau) \approx 1500$ cm^{-1} to stay in accord with the single-crystal data of Refs. 13 and 15.

[18] From the fit of $\sigma_N(\omega,T)$ we obtain $\omega_{pN}/(2\pi c) = 9360$ cm^{-1}, $\gamma = 1/(2\pi c\tau) = 490$ cm^{-1} at 300 K and $\omega_{pN}/(2\pi c) = 7950$ cm^{-1}, $\gamma = 120$ cm^{-1} at 20 K.

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