Direct observation of a nodeless superconducting energy gap in the optical conductivity of iron-pnictides

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The temperature-dependent optical reflectivity and complex transmissivity of an epitaxially grown Ba(Fe0.9Co0.1)2As2 thin film were measured and the optical conductivity and permittivity evaluated over a wide frequency range. The opening of the superconducting gap $2\Delta_0 = 3.7$ meV below $T_c \approx 20$ K is directly observed by a completely vanishing optical conductivity. The temperature and frequency dependent electrodynamical properties of Ba(Fe0.9Co0.1)2As2 in the superconducting state agree well with the BCS predictions with no nodes in the order parameter. The spectral weight of the condensate $1.94 \times 10^7$ cm$^{-2}$ corresponds to a London penetration depth $\lambda_L = 3600$ Å.

I. INTRODUCTION

Soon after the discovery of superconductivity in iron-pnictides,1 the epitaxial growth of LaFeAsO films was reported2 and superconductivity observed in thin films of Co doped SrFe2As2.3 By now the homogeneity of the films and the upper critical field have increased to make iron-pnictides interesting for technological applications.6,7,8 In particular cobalt-doped BaFe2As2 seems to be suitable for producing high-quality thin films which are stable in air9 can be template engineered9 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 electrodynamical properties of the normal and superconducting states.11,12,13 Issues like 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 quasi-particle relaxation are addressed by our optical experiments on Ba(Fe0.9Co0.1)2As2 thin films.

II. EXPERIMENTAL DETAILS AND RESULTS

Ba(Fe1−xCo)x2As2 films were deposited on a (001)-orientated (La,Sr)(Al,Ta)O3 substrate by pulsed laser deposition, where the Ba(Fe0.9Co0.1)2As2 target was ablated with 248 nm KrF radiation under UHV conditions.10 The films grow with a very smooth surface with 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).

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 coefficient amplitude and phase) was measured utilizing a Mach-Zehnder arrangement.15 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)O3 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. For the further analysis we therefore employ a two-layer model that consists of the (La,Sr)(Al,Ta)O3 substrate with thickness of 1.023 mm and optical parameters determined beforehand, covered by the thin film of Ba(Fe0.9Co0.1)2As2. Using Fresnel’s equations,16 we then can analyze the intrinsic optical properties of the film. In the THz range (4 to 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 $\sigma(\omega,T)$.17 The same model was used to evaluate the optical response of the film at higher frequencies. Even-
FIG. 1: (Color online) (a) Reflectivity of a 90 nm Ba(Fe$_{0.9}$Co$_{0.1}$)$_2$As$_2$ film on a 1 nm (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 and phase measurements directly yield the (a) conductivity and (b) permittivity spectra. The dots are directly measured from transmission and phase by a 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$^{-1}$. The inset shows the $T_c = 20$ K.

The properties of the Ba(Fe$_{0.9}$Co$_{0.1}$)$_2$As$_2$ film in the normal state are described by two Drude terms, a narrow $\sigma_N$ and a broad one $\sigma_B$, corresponding to two types of charge carriers as suggested in Ref. 14. In addition, two Lorentz terms account for the interband transitions. In the superconducting state, two additional Drude terms are introduced, one with a tiny scattering rate to model the $\delta$-function (Cooper pair response) obvious in the permittivity spectrum, and another term to describe the quasi-particle contribution to the below-gap conductivity. The optical response at energies around the superconducting gap are mimicked with two Lorentzians since an appropriate expression is missing. The resulting curves $\sigma(\omega,T)$ are plotted in Fig. 1(b) as a function of frequency 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 $T_c$, as depicted in Fig. 2(a): for $T = 5$ K the conductivity has completely vanished. The depletion extends up to approximately 200 cm$^{-1}$. Due to remaining quasi-particles, the conductivity becomes large below 10 cm$^{-1}$ and even exceeds the normal state value. The enormous drop of the low-frequency permittivity $\varepsilon'(\omega,T)$ plotted in Fig. 2(b) evidences the inductive response of the superconducting condensate. It should be noted that the overall conductivity of the Ba(Fe$_{0.9}$Co$_{0.1}$)$_2$As$_2$ film is identical to findings on single crystals 3,15,16 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$_{0.1}$)$_2$As$_2$ has three major components: (i) The infrared peaks at 4400 and 20800 cm$^{-1}$ indicate interband transitions. (ii) The background $\sigma_B \approx 1000$ (Ωcm)$^{-1}$, 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 cannot really be determined. (iii) Most important is the narrow Drude component $\sigma_N(\omega)$, which grows and becomes sharper as $T$ decreases. Its spectral weight decreases by 15% when the temperature is reduced from 300 to 20 K. This corresponds to electronic correlation effects previously observed 17.

For $T < T_c$ the THz conductivity dramatically de-
creases 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. 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. The shape of $\sigma(\omega)$ perfectly agrees with the BCS prediction for a simple s-wave superconductor with no indications of states or nodes in the gap. Previous optical investigations on hole-doped 122 iron-pnictides draw similar conclusions.

Below 10 cm$^{-1}$ a very narrow peak builds up for $T < T_c$ due to the quasi-particle contribution to the conductivity. Its intensity first increases and then diminishes as the quasi-particle number vanishes when $T \rightarrow 0$. 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 ($\nu = 4.6$ cm$^{-1}$) increases strongly right below $T_c$, it passes through a maximum $\sigma_{\text{max}}$ 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 $T < T_c$.

The BCS theory predicts a so-called coherence peak in the low-frequency optical conductivity $\sigma(\omega)$. From the complex transmissivity measurements we can rule out nodes in the gap. The frequency and temperature behavior of the complex electrodynamic response corresponds well with the predictions of the BCS theory.

IV. CONCLUSIONS

The comprehensive optical investigation of Ba(Fe$_{0.95}$Co$_{0.1}$)$_2$As$_2$ thin films evidence the complete opening of the superconducting gap at $2\Delta = 3.7$ meV, i.e. $2\Delta/k_B T_c = 2.1$. From our complex transmissivity measurements we can rule out nodes in the gap. The frequency and temperature behavior of the complex electrodynamic response corresponds well with the predictions of the BCS theory.

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