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Superconducting transition in the Presence of Magnetic order in $BaFe_{1.89}Co_{0.11}As_2$

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Abstract

We report optical spectra of underdoped $BaFe_{1.89}Co_{0.11}As_2$ that hosts both of magnetic and superconducting orders. The temperature dependent evolution of optical conductivity shows finger prints of the magnetic order and the s-wave nature of the superconducting gaps. Careful inspection on the superconducting state reveals that the two orders compete but coexist in the ground state.

Keywords: Superconductivity and magnetism, competing order

1. INTRODUCTION

The unconventional superconductivity in many exotic systems including cuprates and Fe-based superconductors is frequently observed in a vicinity of a magnetic phase, which makes the magnetic interaction at the heart of the discussion about the high temperature superconductivity [1]. The coexistence of the magnetism and superconductivity over a broad range of a phase diagram of Fe-based superconductors provides an ideal testbed for investigation of their interplay. In Fe-based superconductors doping or pressure suppress the magnetic order while the superconductivity develops. Therefore, it is suspected that the static magnetic order competes against the superconductivity. Indeed, there have been indications that the magnetic order is suppressed when it enters to the superconducting state [2-4]. Sometimes, however, the magnetic order is detected only when the sample is cooled down below the superconducting transition temperature, which suggests that a magnetic order can cooperate with the superconductivity [4].

In this work, we investigate the optical spectra of underdoped $BaFe_{1.89}Co_{0.11}As_2$ that shows a structural transition at $T_s \sim 50$ K, the magnetic order at $T_m \sim 32$ K, and the superconducting transition at $T_c \sim 21.7$ K. We observe clear signature of the magnetic order that starts to develop already below T_s. When the sample is cooled below T_c, superconducting gaps of s-wave nature open. The observed temperature dependence is different from that of conventional superconductors, where no competing order exists. We demonstrate that the magnetic order partly competes with the superconductivity but still coexists with it based on conductivity variation and the reflectivity ratio between superconducting and normal states.

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2. EXPERIMENT

Single crystals of $BaFe_{1.89}Co_{0.11}As_2$ were grown from self-flux in glassy carbon crucibles and their chemical composition was determined by energy dispersive X-ray spectroscopy as described in [5]. Resistivity and dc magnetization measurements show a sharp SC transition at T_c =21.7 K [2]. Infrared spectroscopy measurements were performed on ab-plane surfaces of freshly cleaved crystals from the same growth batch. The reflection and ellipsometry techniques for the optical measurements are described in [6]. We performed Kramers-Kronig (KK) analysis to obtain optical conductivity spectra $\sigma(\omega)$ from the reflectivity data. The Hagen-Rubens relation was adopted for the low frequency extrapolation below 30 cm^{-1} [7]. Because the extrapolations with the measured resistivity do not match the measured reflectivity, the resistivity values are scaled by a constant factor for all temperatures such that the Hagen-Rubens extrapolations connect smoothly to the measured reflectivity spectra. We note that the KK transformed spectra do not depend significantly on the resistivity values used for the extrapolation except in the low frequency limit below 30 cm^{-1}.

3. RESULTS AND DISCUSSION

Fig. 1 shows the measured reflectivity in the far infrared region and conductivity spectra of $BaFe_{1.89}Co_{0.11}As_2$ at

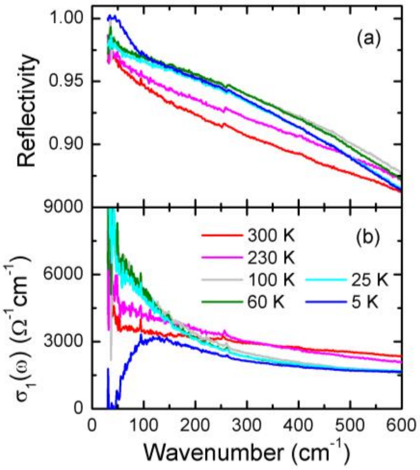

Fig. 1. Temperature dependent (a) reflectivity and (b) optical conductivity in the far infrared region. As temperature is cooled below T_c, reflectivity approaches 1 and the superconducting gap opens.

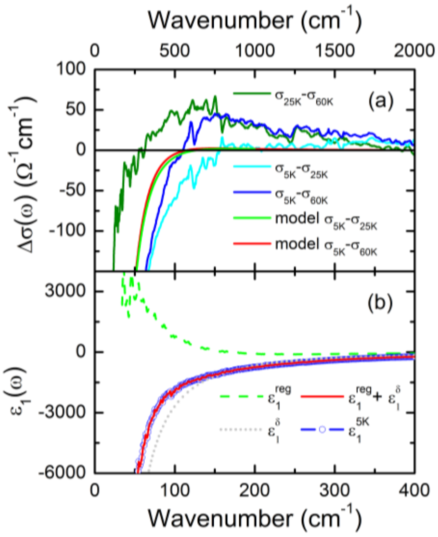

Fig. 2. (a) The change of the optical conductivity $\Delta\sigma(\omega)$ across the magnetic and superconducting transitions. The variation of the model calculation between 2- and 3-gap models is smaller than the line thickness and the 2-gap cases are plotted. (b) Proof of the delta function response. The regular part contribution ε_1^{reg} is obtained by KK transformation of $\sigma(\omega)$ at 5 K. The sum of ε_1^{reg} and the delta function response ε_1^{δ} agrees with the measured data at 5 K.

selected temperatures. At high temperatures above 60 K, the overall reflectivity in the far infrared region increases as the temperature decreases. It makes the conductivity increase below 200 cm^{-1} but decrease above 400 cm^{-1}. This behavior could be related with the decrease of the scattering rate and/or temperature dependent modification of the correlation effect that might open a pseudogap [8]. As the material is further cooled below 50 K, reflectivity at higher frequency decreases significantly, which originates from the magnetic order. Although the spin density waves of the undoped compound $BaFe_2As_2$ introduces a strong gap signature in the conductivity, the corresponding behavior does not appear clearly in Fig. 1(b). However, the gap-like feature due to the magnetic order in $BaFe_{1.89}Co_{0.11}As_2$ manifests itself in the difference spectra of the conductivity as shown in Fig. 2(a). We note that although the static magnetic order is stabilized below $T_m \sim 32$ K the gap-like feature develops already below T_s because of the fluctuating order [2].

When the temperature is lowered below T_c, the reflectivity in the low frequency below 100 cm^{-1} suddenly increases and approaches 1 at about 45 cm^{-1} at 5K, while higher frequency response is relatively frozen. Such a behavior is typical for s-wave superconductors, which becomes evident in the conductivity spectrum. That is, a superconducting gap of 2Δ about 47 cm^{-1} can be deduced from Fig. 1(b) where the conductivity is suppressed to zero. Fig. 2(b) confirms that the energy gap seen in $\sigma(\omega)$ indeed originates from a superfluid. It shows that the dielectric function $\varepsilon_1(\omega)$ at 5 K is the sum of the delta function response ε_1^{δ} and ε_1^{reg} of the regular part that does not condense but shows up in $\sigma(\omega)$. The sum rule requires that the weight of the superfluid should be the same with the integrated spectral weight of the suppressed part $\Delta\sigma(\omega) = \sigma_{25K}(\omega) - \sigma_{5K}(\omega)$ [7]. We find that the two quantities agree nicely with each other within 3 % of the estimated value $\omega_p^2 \sim 2.7\times10^7$ cm^{-2}. It is worth to note that the conductivity at low frequency is subject to a large error depending on the extrapolation and the dielectric function should provide the better estimation of the weight of the superconducting delta function.

It is known that the Fe-based materials host multiple superconducting energy gaps and a single gap model fails to explain the optical spectra [6, 9, 10]. Although some materials present clear signatures from different superconducting gaps, the kinks from higher energy gaps are usually blurred and a careful inspection is needed to obtain further details about the gaps. Therefore, it is challenging to distinguish a gap with a relatively small spectral weight. In underdoped Fe-based superconductors, the situation gets further complicated due to the presence of the magnetic order that may compete with the superconductivity.

Fig. 3 shows a few trial fits to the 5 K spectra with various conditions to understand the superconducting state. We employ Drude and Lorentz oscillator models for the normal states and adopt BCS type superfluid responses given in [11]. The superconducting response is obtained by imposing superconducting energy gaps to the Drude terms that describe the normal state. The far infrared free carrier response above 30 cm^{-1} at 25 K is modeled with two Drude terms of $\gamma_1 = 135$ cm^{-1} and $\gamma_2 = 450$ cm^{-1}. We note that the Hagen-Rubens extrapolation with scaled resistivity introduces a very narrow spectral feature in the low frequency below 30 cm^{-1}. Therefore, another Drude term with $\gamma_3 = 15$ cm^{-1} is added to account for the feature in the extrapolation range, which does not modify significantly the higher frequency response above 30 cm^{-1}. Because the damping $\gamma_3 = 15$ cm^{-1} is small enough not to contribute to

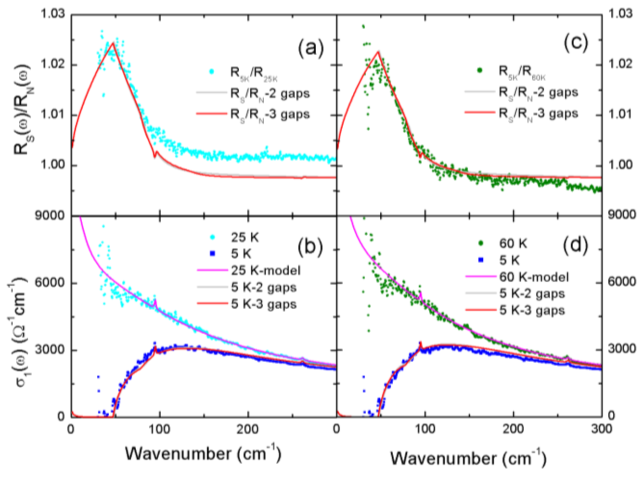

Fig. 3. Model fits to the normal and superconducting states. (a), (b) show the fits considering the 25 K spectra as the normal state response and (c), (d) show those considering 60 K spectra as the normal state ones.

the 5 K spectra above the full gap of $2\Delta \sim 47$ cm^{-1}, this Drude term hardly influences the far infrared behavior in the superconducting state except for adding its full weight to the delta function. In the following, we discuss parameters of two broader Drude terms that determine the far infrared spectra above 30 cm^{-1}. In the 2-gap model, two gap values of $2\Delta = 48$ cm^{-1}, 82 cm^{-1} are assumed for the two Drude terms respectively. In the 3-gap model, gaps of $2\Delta = 47$ cm^{-1}, 138 cm^{-1} are supposed to share the weight of one Drude term with $\gamma_1 = 135$ cm^{-1} and $2\Delta = 78$ cm^{-1} is assigned to the other.

As shown in Fig. 3(a) and Fig. 3(b), the fitting qualities of the two models are not significantly different from each other. Without a clear feature for the third gap, it may not need to discuss with three gaps. One can notice that there is noticeable disagreement in the fits of $\sigma(\omega)$. Most important observation is that all model fits never reproduce a correct level of the reflectivity ratio R_{5K}/R_{25K} above the gap. Here, we note that the R_S/R_N value of the model fit does not depend on the choice of parameters for the normal state spectra at 25 K. It is worth to mention that a mode is desired to be included to account for the magnetic order at 25 K although its weight is small [2]. Unfortunately there is no theoretical model function to fit the observed feature of $\Delta\sigma(\omega)$ in Fig. 2. Instead, different Drude terms with additional Lorentz oscillators could be used for the magnetic state. However, all those cases fail to meet the observed level of R_{5K}/R_{25K} above the gap as long as the superconducting gaps are simply imposed to the Drude terms at 25 K. We note that R_S/R_N becomes always slightly smaller than 1 right above the largest superconducting gap and approaches 1 in the higher frequency when superconductivity develops in Drude terms without modifying other modes.

In most superconductors, R_S/R_N has been shown close to 1 not only in conventional superconductors but also unconventional materials including cuprates and optimally doped Fe-based superconductors [6], [12-15]. However, in many of the cases the experimental error bar is too large to discuss the behavior observed in this work. Interestingly, there is a report of the reflectivity change across a superconducting transition even in the visible region beyond 2 eV in $Tl_2Ba_2Ca_2Cu_3O_{10}$ [16]. They argued that the paring is mediated by the electronic mode in the visible region, which may result in the reflectivity change over a wide range with R_S/R_N larger than 1 up to 1 eV. Reference [17] pointed out that the temperature dependent evolution of scattering and correlation effects should be considered carefully. Abrupt spectral changes in the visible region across superconducting and magnetic phase transitions are reported also in Fe-based materials [18]. These discussions suggest that the electronic structure could be strongly modified when the sample enters to the superconducting state and it may result in R_S/R_N larger than 1.

The answer to the failure of the model fits of Fig. 3(a) and Fig. 3(b) can be found in the spectrum R_{5K}/R_{60K}. Although the normal state fit is to the spectra at 25 K in Fig. 3(a) and Fig. 3(b), model calculation of R_S/R_N right above the gaps gives always the value of R_{5K}/R_{60K}. It has been reported that although the magnetic order coexists with the superconductivity in the underdoped region, they tend to compete for the spectral weight [2]. Indeed, model calculations of the superconducting response assuming 60 K as the normal state shown in Fig. 3(c) and Fig. 3(d) agree better with the experiments. We stress that the level of R_S/R_N above gaps does not depend on the details of the fits as long as the overall gap magnitudes and weights of superconducting and normal states were not altered significantly. This result implies that the dominant feature of the superconducting state develops not from 25 K with the magnetic order but from 60 K without the magnetic order.

However, it should be noted that the model calculation does not fully agree with R_{5K}/R_{60K}. That is, R_{5K}/R_{60K} decreases strongly as frequency increase beyond 300 cm^{-1} that can be noticed in Fig. 1(a), while the model expects R_S/R_N to increase to 1. It suggests that the response at 60 K does not fully represent the normal state response either from which the superconductivity develops. The difference spectrum of $\Delta\sigma(\omega) = \sigma_{60K}(\omega) - \sigma_{5K}(\omega)$ indeed suggests that although the magnetic order is suppressed in the superconducting state at 5 K in the low frequency region, it still persists above 500 cm^{-1} while all model calculations expect nearly no change of $\Delta\sigma(\omega)$ above 500 cm^{-1}. The remaining feature above 500 cm^{-1} should influence the higher frequency response in the superconducting state. It suggests that the magnetic order is suppressed in the superconducting state but partly survives together with the superconductivity. We remark that following two factors should be also considered carefully. Although the temperature is already very low, the overall scattering rate of the system could be modified across the phase transitions and some spectral weight may be added in the low frequency region as suggested in [18]. Thorough understanding of the subtle change of electronic structure will unveil the pathway to the high temperature superconductivity.

4. CONCLUSION

We observed clear signatures of the magnetic order and s-wave superconducting gap in $BaFe_{1.89}Co_{0.11}As_2$. The conductivity spectrum in the superconducting state can be well described by a model with BCS type energy gaps. However, the reflectivity ratio across the superconducting transition suggests that the normal state from which the superconductivity develops is not the one just above T_c nor the one without the magnetic order. The difference spectra suggest that the magnetic order is partly suppressed when the superconductivity develops but it continues to influence the electronic structure in the ground state.

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REFERENCES

[1] D. N. Basov and Andrey V. Chubukov, “Manifesto for a higher T_c,” *Nat. Phys.*, vol. 7, p. 272, 2011.

[2] P. Marsik *et al*., “Coexistence and competition of magnetism and superconductivity on the nanometer scale in underdoped $BaFe_{1.89}Co_{0.11}As_2$,” *Phys. Rev. Lett*., vol. 105, p. 057001, 2010.

[3] J. Larsen *et al*., “Competing superconducting and magnetic order parameters and field-induced magnetism in electron-doped $Ba(Fe_{1-x}Co_x)_2As_2$,” *Phys. Rev. B*, vol. 91, p. 024504, 2015.

[4] C. Bernhard *et al*., “Muon spin rotation study of magnetism and superconductivity in $Ba(Fe_{1-x}Co_x)_2As_2$ single crystals,” *Phys. Rev. B*, vol. 86, p. 184509, 2012.

[5] F. Hardy, P. Adelmann, T. Wolf, H. v. Lohneysen, and C. Meingast, *Phys. Rev. Lett*., vol. 102, p. 187004, 2009.

[6] K.W. Kim *et al*., “Evidence for multiple superconducting gaps in optimally doped $BaFe_{1.87}Co_{0.13}As_2$ from infrared spectroscopy,” *Phys. Rev. B* vol. 81, p. 214508, June 2010.

[7] M. Dressel and G. Grüner, *Electrodynamics of solids: optical properties of electrons in matter*, Cambridge University Press, 2002.

[8] S. J. Moon *et al*., “Infrared pseudogap in cuprate and pnictide high-temperature superconductors,” *Phys. Rev. B*, vol. 90, p. 014503, 2014.

[9] E. van Heumen *et al*., “Optical properties of $BaFe_{2-x}Co_xAs_2$,” *Euro. Phys. Lett*., vol. 90, p. 37005, 2010.

[10] C. Homes *et al*., “Electronic correlations and unusual superconducting response in the optical properties of the iron-chalcogenide FeTe0.55S0.45,” *Phys. Rev. B*, vol. 81, p. 180508(R), 2010.

[11] W. Zimmermann, E. H. Brandt, M. Bauer, E. Seider, and L. Genzel, “Optical conductivity of BCS superconductors with arbitrary purity,” *Physica C*, vol. 183, pp. 99-104, 1991.

[12] G. Li *et al*., “Probing the superconducting energy gap from infrared spectroscopy on a $Ba_{0.6}K_{0.4}Fe_2As_2$ single crystal with T_c = 37 K,” *Phys. Rev. Lett.*, vol. 101, p. 107004, 2008.

[13] K. E. Kornelsen, M. Dressel, and J. E. Eldrige, “Far-infrared optical absorption and reflectivity of a superconducting NbN film,” *Phys. Rev. B*, vol. 44, pp. 11882-11887, 1991.

[14] Z. Schlesinger, R. T. Collins, M. W. Shafer, and E. M Engler, “Normal-state reflectivity and superconducting energy gap measurement of $La_{2-x}Sr_xCuO_4$,” *Phys. Rev. B*, vol. 36, pp. 5275-5278, 1987.

[15] U. Nagel *et al*., “Far-infrared signature of the superconducting gap in intercalated graphite CaC_6,” *Phys. Rev. B*, vol. 78, p. 041404(R), 2008.

[16] M. J. Holcomb, J. P. Collman, and W. A. Little, “Optical evidence of an electronic contribution to the pairing interaction in superconducting $Tl_2Ba_2Ca_2Cu_3O_{10}$,” *Phys. Rev. Lett.*, vol. 73, pp. 2360-2363, 1994.

[17] A. V. Boris *et al.*, “In-plane spectral weight shift of charge carriers in $YBa_2Cu_3O_{6.9}$,” *Science*, vol. 304, pp. 708-710, April 2004.

[18] A. Charnukha *et al*., “Superconductivity-induced optical anomaly in an iron arsenide,” *Nat. Commun.*, vol. 2, p. 219, 2011.