Eliashberg Analysis of Optical Spectra Reveals Strong Coupling of Charge Carriers to Spin Fluctuations in Superconducting Iron Pnictides

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The temperature and frequency dependences of the optical conductivity of Co and Ni-doped BaFe$_2$As$_2$ are analyzed and the electron-boson spectral density $\alpha^2 F(\omega)$ extracted using Eliashberg’s formalism. The characteristic energy of a large peak in the spectrum around 10 meV coincide with the resonance peak in the spin excitation spectra, giving compelling evidence that in iron-based superconductors spin fluctuations strongly couple to the charge carriers and mediate superconductivity. In addition the spectrum is found to evolve with temperature towards a less structured background at higher energies as in the spin susceptibility.

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Whenever a new family of superconductors is discovered, among the first questions posed are about the mechanism of superconductivity. In conventional superconductors, phonons mediate the attractive interaction of two electrons forming Cooper pairs. For heavy fermions, cuprates and some organic superconductors, magnetic Cooper-pairing mechanisms have been proposed as a candidate. However, intrinsic complications have prevented the general acceptance. The situation of the novel class of iron based superconductors is comparably unsettled. Nevertheless, the proximity of superconductivity and antiferromagnetism in the phase diagram, the weak electron-phonon interaction and the resonance peak in the spin-excitation spectrum support the hypothesis of a magnetic interaction leading to superconductivity.

The Eliashberg spectral function $\alpha^2 F(\omega)$ quantifies the boson exchange effect and is a good way to discriminate between the candidate mechanisms. For half a century, current-voltage characteristics obtained by tunneling spectroscopy are utilized to provide detailed and accurate information on the phonon exchange for most conventional superconductors. In strongly correlated systems, such as high-$T_c$ cuprates, the inversion of optical data is commonly used to extract the bosonic excitation spectra. The analysis and interpretation, however, is not straightforward since a certain complexity in understanding the spectral signature of bosonic modes arises from the joint mechanisms in these particular systems. In this regard, Carbotte and collaborators succeeded to modify the kernel $\alpha^2 F(\omega)$ of the Eliashberg theory by introducing the nearly antiferromagnetic Fermi-liquid model where the exchanged bosons are described as antiferromagnetic spin fluctuations, and the resulted optical resonance tracked very well the temperature evolution of the spin resonance seen in neutron scattering.

In this Letter, we reported a detailed analysis of our optical spectra obtained on Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$ single crystals. The inversion of the frequency dependent optical scattering rate $\tau^{-1}(\omega)$ reveals that the coupling of charge carriers to bosonic modes has an optimum peak around 10 meV, with a coupling constant $\lambda = 4.4$ right above $T_c$. With increasing temperature, this peak becomes broader and moves to higher energies. These bosonic spectral signatures closely resemble the dynamical spin susceptibility $\chi'(\omega)$ observed by neutron scattering, indicating a magnetic mediation mechanism in the novel iron-pnictide superconductors.

We have measured the optical reflectivity of Co-doped BaFe$_2$As$_2$ single crystals over a wide frequency and temperature range as described in detail in Ref. 18. The samples are well characterized and exhibit a superconducting transition at $T_c = 25$ K. Via Kramers-Kronig analysis we calculate the complex conductivity $\tilde{\sigma} = \sigma_1 + i\sigma_2$ which is further analyzed by the extended Drude model in order to obtain the frequency dependent scattering rate $1/\tau(\omega)$ and mass enhancement $m^*(\omega)/m_0 = 1 + \lambda(\omega)$ compared to the bandmass $m_0$. The results are plotted in Fig. 1 for different temperatures.

In principle, optical data are encoded with information on the microscopic interaction between the charge
the form

\[
\frac{1}{\tau(\omega)} = \frac{1}{\tau_{\text{imp}}} + \int_0^\infty K(\omega, \Omega; T) \alpha^2 F(\Omega) \, d\Omega ,
\]

where \(1/\tau_{\text{imp}}\) denotes a constant scattering rate due to impurities; the normal state kernel is given by \([21]\):

\[
K(\omega, \Omega; T) = \frac{\pi}{\omega} \left\{ \frac{h\Omega}{2k_B T} \right\} - (\omega - \Omega) \coth \left( \frac{\omega + \Omega}{2k_B T} \right) + (\omega - \Omega) \coth \left( \frac{\omega - \Omega}{2k_B T} \right).
\]

Several methods have been suggested to extract the information on the electron-boson spectral density \(\alpha^2 F(\omega)\) from the optical scattering rate, such as singular value decomposition, maximum entropy method and least square fit; a detailed discussion of advantages and limitations of these numerical inversion techniques is given in Ref. \([22]\). Here we have approached the deconvolution by an unbiased maximum entropy method, similar to \([17]\), and plot the outcome in Fig. 1(c) for various temperatures in the metallic state of Ba(Fe\(_{0.92}\)Co\(_{0.08}\))\(_2\)\(\text{As}_2\).

Note, when a gap in the density of states opens below \(T_c\), the present analysis of \(\tau^{-1}(\omega)\) becomes meaningless; hence we have to restrict ourselves to \(T > T_c\). However, it is safe to assume that electron-boson coupling makes a strong impact on the spectra already in the normal state just above \(T_c\), and it is this quantity which will determine \(T_c\). Before starting the discussion, we would like to emphasize that the Eliashberg inversion applied here is based on single band systems with infinite band width while most of materials actually have finite band width. The iron pnictides, on the other hand, are certainly multiband systems (see for instance Refs. \([10]\) and \([12]\)). In the normal state the optical conductivity of such a multi-band system is just the sum of the various band contributions to this conductivity, provided the interband optical transitions are zero or sufficiently small, as was suggested by van Heumen \textit{et al.} \([23]\) for the Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)\(\text{As}_2\) class of materials. As is generally believed, the inelastic scattering is dominated by interband transitions due to possibly spin fluctuations and, thus, a single form of the electron-boson spectral density will enter the problem except for a possible scaling factor accounting for a different magnitude of \(\alpha^2 F(\omega)\) for transitions between different bands. This was considered explicitly by Benfatto \textit{et al.} \([24]\). Thus, the application of Eq. (1) for the deconvolution of experimental data will yield meaningful information about the electron-boson interaction in such systems and will provide an average \(\alpha^2 F(\omega)\) spectrum. The shape of such a spectrum will still provide meaningful information on the bosons responsible for superconductivity, for example phonons or spin fluctuations. A knowledge of its average magnitude is also equally important. Finally, the use of a formula based on infinite band width to deconvolute optical data.

FIG. 1: (Color online) Results of the inversion calculations of the electron-boson spectral function \(\alpha^2 F(\omega)\) for Ba(Fe\(_{0.92}\)Co\(_{0.08}\))\(_2\)\(\text{As}_2\) at different temperatures in the normal state. (a) Measured conductivity spectra (solid lines) compared with computational results (dashed lines). (b) Frequency dependent scattering rate obtained by the extended Drude analysis of the conductivity spectra plotted in the upper panel (solid lines) compared with the calculated \(1/\tau(\omega)\) according to Eq. (1) (dashed lines) with a constant contribution of 304 cm\(^{-1}\) due to impurities. (c) The corresponding electron boson spectral density \(\alpha^2 F(\omega)\) calculated by Eliashberg formalism.

Carriers. For an electron-boson system, the Eliashberg equations apply and a Kubo formula can be used to get the infrared conductivity from the electron-boson spectral density once the quasiparticle self-energy \(\Sigma(\omega; T)\) is known \([20]\). The opposite direction turns out to be even more challenging since we have to invert an equation of
of systems with a rather narrow band width makes it extremely important to check the Kramer-Kronig consistency of $\alpha^2 F(\omega)$ with optical constants to exclude any incorrect solutions due to finite band-width effects. In order to demonstrate the applicability of our analysis, we added the calculated $\sigma_1(\omega)$ and $\tau^{-1}(\omega)$ to Fig. 1(a) and (b) as dashed lines; both cases show good agreement between theory and experiment. This gives confidence in the physical relevance of the derived spectra.

In Fig. 1(c), one sees a clear temperature dependence to the recovered electron-boson spectra which is also in agreement with a second, biased maximum entropy inversion [17]. At temperatures just above $T_c$, a pronounced peak centered at 10 meV and a shoulder around 45 meV dominate the spectral weight below 80 meV. When $T$ increases, this peak moves to higher energies and smears out quickly as a shoulder. As a consequence, the mass renormalization factor $\lambda$ is reduced from 4.4 to 1.67 at $T = 200$ K. Since 10 meV seems to be a reasonable energy for phonon excitations, at first glance, it is tempting to consider a phonon mechanism for superconductivity. However, compared to band structure calculations [6, 7], the observed spectral features are quite different. Moreover, from calculations of the electron-boson excitation spectra a characteristic phonon frequency $\omega_{in}$ can be extracted; in our case $\hbar \omega_{in} = 14.2$ meV. When the phonon mechanism is dominant in a superconducting material, one can estimate the coupling strength by its ratio to $T_c$.

Here, we obtain $k_B T_c/\hbar \omega_{in} = 0.15 < 0.25$, implying a conventional strong coupling material; it also yields $\lambda$ to be in the range $1 - 2$, according to the McMillan equation [13]. Obviously, this is much too small compared to our experimental result ($\lambda = 4.4$). Here, we would like to point out that the mass renormalization factor $\lambda$ is widely reported to be $\lambda = 4-5$ by other studies [25]. Thus, we expect another mechanism to play the key role in mediating superconductivity in these materials.

As mentioned above, spin fluctuations seem to be the natural candidate for the superconducting “glue” in iron-based materials. Whenever a magnetic mechanism is discussed, the main concern is whether spin-fluctuation exchange provides sufficient spectral intensity to make a significant impact on the electronic self-energy. Very recently, Dalm et al. [20] succeed to establish a quantitative relationship between the charge- and spin-excitation spectra in high-$T_c$ cuprates, which demonstrates that the magnetic interaction can generate $d$-wave superconducting states with transition temperatures comparable to the maximum $T_c$ observed in these compounds; in other words, spin fluctuations do have enough strength to cause superconducting transitions at high-temperature. In Fig. 2 we display the electron-boson spectral function $\alpha^2 F(\omega)$ derived from optical scattering rate of Ba-$(\text{Fe}_{0.92}\text{Co}_{0.08})_2\text{As}_2$ together with the spin excitation spectrum $\chi'(\omega)/\chi(\omega)$ of Ba$(\text{Fe}_{0.92}\text{Co}_{0.075})_2\text{As}_2$ obtained from inelastic neutron scattering experiment [8]. Both spectra show a resonance at approximately 10 meV at low temperature, and the peak smears out when the temperature increases. Such an important agreement between optics and neutron indicates that the charge carriers in this material are strongly coupled to the spin excitations.

Our results for Ba$(\text{Fe}_{0.95}\text{Ni}_{0.05})_2\text{As}_2$ ($T_c = 20$ K) are qualitatively similar, but span only a limited energy range and exhibit excessive noise due to the smaller crystal size [18, 19]. Inelastic neutron scattering indicates a resonance peak in the spin excitation spectrum around 7 meV [9] corresponding to the lower energy scale in this material. Yang et al. [27] performed a similar analysis on K doped BaFe$_2$As$_2$ and found two maxima of $\alpha^2 F(\omega)$ in the range below 30 meV. Although different in detail, the overall accord gives us confidence that our observations reveal a general behavior in this class of materials.

In conclusion, we have analyzed the temperature and frequency dependencies of the optical properties of doped BaFe$_2$As$_2$ via Eliashberg theory. We obtained the electron-boson spectral density $\alpha^2 F(\omega)$ which exhibit a characteristic peak around 10 meV. This coincides with the resonance peak in the spin excitation spectrum and gives evidence that in iron-based superconductors spin fluctuations strongly couple to the charge carriers and mediate superconductivity. Also there is a strong evolution of the spectra with temperature which agrees with the known spin fluctuation spectra and this would not be the case in a phonon mechanism for which the $\alpha^2 F(\omega)$ is expected to remain independent of temperature.

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