Effective medium approximation and the complex optical properties of the inhomogeneous superconductor $K_{0.8}Fe_{2-y}Se_2$

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The in-plane optical properties of the inhomogeneous iron-chalcogenide superconductor $K_{0.8}Fe_{2-y}Se_2$ with a critical temperature $T_c = 31$ K have been modeled in the normal state using the Bruggeman effective medium approximation for metallic inclusions in an insulating matrix. The volume fraction for the inclusions is estimated to be $\geq 10\%$; however, they appear to be highly distorted, suggesting a filamentary network of conducting regions joined through weak links. The value for the plasma frequency $\omega_{p,D}$ in the inclusions is much larger than the volume average, which when considered with the reasonably low values for the scattering rate $1/\tau_D$, suggests that the transport in the grains is always metallic. Estimates for the dc conductivity $\sigma_L$ and the superfluid density $\rho_s$ in the grains place the inclusions on the universal scaling line $\rho_s/8 \simeq 4.4\sigma_L T_c$ close to other homogeneous iron-based superconductors.

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I. INTRODUCTION

The surprising discovery of superconductivity in the iron-chalcogenide compounds with relatively high critical temperatures ($T_c$’s) has generated a great deal of interest in these materials. The minimal electronic structure is characterized by hole and electron pockets at the center and corners of the Brillouin zone, respectively. It has been proposed that the scattering between the electron and the hole pockets forms the basis of a spin-fluctuation pairing mechanism. For this reason, the discovery of superconductivity in $K_{0.8}Fe_{2-y}Se_2$ was of particular interest because the hole pocket in this material was absent, yet a relatively high $T_c \simeq 31$ K was observed, as opposed to the hole-doped analog $KFe_2As_2$, which had a dramatically reduced $T_c \simeq 3$ K. This suggests that the pairing mechanism may not be a settled issue in these materials. A further complication in understanding the physical properties of $K_{0.8}Fe_{2-y}Se_2$ arises from the growing body of evidence that suggests that this material is not homogeneous but, instead, consists of nonmagnetic metallic (superconducting) inclusions in a magnetic insulating matrix. The optical properties of $K_{0.8}Fe_{2-y}Se_2$ and the related $Rb_yFe_xSe_5$ material have been investigated in some detail and support the conclusion that these materials are inhomogeneous. The phase-separated nature of these materials complicates the optical determination of the complex dielectric function, which is, by nature, a volume-averaging technique. However, a recent study of $K_{0.8}Fe_{2-y}Se_2$ by Wang et al. noted that the optical properties of this material could be described quite well using an effective medium theory for the dielectric function, which consists of separate contributions from metallic inclusions embedded in an insulating matrix. Our original paper on the optical properties of $K_{0.8}Fe_{2-y}Se_2$ noted that the normal- and superconducting-state properties both indicated that this material was inhomogeneous and that the superconductivity was due to Josephson coupling between the superconducting regions. In view of the phase-separated nature of this material, the application of an effective medium theory to our optical data is a necessary next step in modeling the optical properties. In this paper, we apply the Bruggeman effective medium approximation dielectric function to the normal-state optical properties of $K_{0.8}Fe_{2-y}Se_2$. The metallic inclusions appear to comprise about 10% of the total sample volume, resulting in a Drude plasma frequency that is significantly higher than the volume-averaged value but is still much smaller that the values observed in other (homogeneous) iron-based superconductors, unless volume fractions of less than 1% are considered. Interestingly, and in agreement with another recent paper on the optical properties of this material, the effective medium approximation (EMA) cannot be successfully applied to the data without assuming that the inclusions are extremely distorted, suggesting the formation of filamentary conducting networks. The estimated superconducting plasma frequency of the inclusions is again much larger than the volume-averaged value but is still significantly smaller than the values determined in other iron-based superconductors. The volume-averaged values for the dc conductivity (measured just above $T_c$) and the superfluid density placed this material on the universal scaling for the cuprate materials, albeit in a region associated with Josephson coupling. In contrast, the inferred superfluid density of the metallic (superconducting) inclusions falls on the same scaling line but in a region associated with coherent transport and conventional superconductivity.

II. METHOD

There are two general theories for an effective medium. The first is the Maxwell-Garnet dielectric function, which considers a dilute system of inclusions. The difficulty with this approach is that it is asymmetric with respect to the inclusions and the matrix. The second is the Bruggeman EMA, which is symmetric with respect to the inclusions and the matrix and is not restricted to any particular range of concentrations. Another advantage of the EMA dielectric function is that it correctly predicts the percolation threshold for spherical grains. Because the metallic inclusions may represent a large volume of the sample, we have chosen the Bruggeman EMA dielectric function. For inclusions with complex dielectric
Photon energy (meV)

Wave number (cm⁻¹)

![Image](144530-2)

FIG. 1. (Color online) The absolute reflectance over a wide frequency range for a cleaved single crystal of K₀.₈Fe₂₋₄Se₂ for light polarized in the α-β planes at several temperatures above and below Tₙ. Inset: The temperature dependence of the real part of the dielectric function in the far-infrared region.

function ˜εᵦ with a volume fraction f in a matrix ˜εᵦ, the EMA dielectric function ˜ε is the root of the quadratic expression,²⁰⁻³²

\[ f \frac{\tilde{\epsilon}_a - \tilde{\epsilon}}{\tilde{\epsilon}_a + \phi_c \tilde{\epsilon}} + (1 - f) \frac{\tilde{\epsilon}_b - \tilde{\epsilon}}{\tilde{\epsilon}_b + \phi_c \tilde{\epsilon}} = 0, \]

where the physical solution is the one that has Im(˜ε) > 0. Here, \( \phi_c = (1 - g_c)/g_c \), where \( g_c \) is the depolarization factor for a spheroid,

\[ g_c = \frac{1 - e_c^2}{e_c^2} \left[ \frac{1}{e_c \tanh(e_c)} - 1 \right]. \]

For a spheroid with figure axis length a and transverse axis length b, the eccentricity of a spheroid is \( e_c = \sqrt{1 - a^2/c^2} \).

The EMA dielectric function is fit to the reflectance using a nonlinear least-squares approach; the reflectance is chosen because it is a combination of the both the real and the imaginary parts of the dielectric function, as opposed to the real part of the conductivity, which depends only on the imaginary part of ˜ε. The temperature dependence of the reflectance for light polarized in the planes of a single crystal of K₀.₈Fe₂₋₄Se₂ with \( Tₙ = 31 \) K was determined in a previous paper²⁸ and was reproduced in Fig. 1 with the real part of the dielectric function shown in the inset. Despite being a volume-averaged measurement, at low temperatures, the real part of the dielectric function falls below zero at low frequencies, indicating a weakly metallic state. The inclusions are assumed to be metallic in the normal state (superconducting below \( Tₙ \)) with a complex dielectric function that may be described by a simple Drude model,

\[ \tilde{\epsilon}_d(\omega) = \epsilon_∞ - \frac{\omega_p,D^2}{\omega^2 + i\omega/\tau_D}, \]

where \( \epsilon_∞ \) is the real part of the dielectric function at high frequencies, \( \omega_p,D^2 = 4\pi n e^2/m^* \) and \( 1/\tau_D \) are the square of the plasma frequency and scattering rate for the delocalized (Drude) carriers, respectively, and \( m^* \) is an effective mass. The matrix is assumed to be insulating with a complex dielectric function consisting only of Lorentz oscillators,

\[ \tilde{\epsilon}_β(\omega) = \epsilon_∞ + \sum_j \frac{\Omega_j^2}{\omega^2 - \omega_j^2 - i\omega\gamma_j}. \]

where \( \omega_j, \gamma_j, \) and \( \Omega_j \) are the position, width, and oscillator strength of the jth vibration. In addition to \( \omega_p,D \) and \( 1/\tau_D \) in \( \tilde{\epsilon}_d \) and the stronger oscillators in \( \tilde{\epsilon}_β \), f and \( \phi_c \) are both allowed to vary; \( \epsilon_∞ \) is also fit but is assumed to be the same in both \( \tilde{\epsilon}_d \) and \( \tilde{\epsilon}_β \). (It should be noted that, in a metallic system, the optical properties at low frequencies are largely independent of \( \epsilon_∞ \).) From the EMA dielectric function, at normal incidence, \( \tilde{\epsilon} = (\sqrt{\epsilon - 1})/(\sqrt{\epsilon + 1}) \) and \( R = \tilde{\epsilon}^2 \). The complex conductivity is \( \tilde{\sigma}(\omega) = \sigma_1 + i\sigma_2 = -i\omega[\tilde{\epsilon}(\omega) - \epsilon_∞]/4\pi \).

The results of the nonlinear least-squares fits of the EMA dielectric function to the normal-state reflectance at 200, 100, and 35 K are shown in Fig. 2 as well as a comparison of the experimentally determined optical conductivity with the calculated EMA result; the agreement with the experiment is excellent. It has been previously remarked that the quality of the EMA fits is contingent on the size of the metallic inclusions being much smaller than the wavelength of the radiation in the solid.²⁵ Given the average index of refraction \( n \approx 4 \) in this material²⁸ and the frequency interval of the fit, a very rough value of the size of the inclusions is that they are no larger...
III. RESULTS AND DISCUSSION

A. Volume fraction

The EMA fit to the reflectance at 35 K is shown in Fig. 2(a) with the comparison to the conductivity shown in the inset; the fitted parameters are \( f \approx 0.11 \) and \( \phi_c \approx 30 \) with the Drude parameters \( \omega_{p,D} \approx 1320 \) and \( 1/\tau_D \approx 57 \text{ cm}^{-1} \). For the fits at 100 and 200 K shown in Figs. 2(b) and 2(c), \( f, \phi_c, \) and \( \omega_{p,D} \) are fixed, and the scattering rate is allowed to vary, returning \( 1/\tau_D \approx 126 \) and 144 \text{ cm}^{-1} \), respectively. (Note that the optical properties at 200 and 295 K are almost identical.)

The estimate that only about 10% of the sample is conducting (superconducting) is in good agreement with recent Mössbauer, NMR, and bulk muon-spin rotation (\( \mu \)SR) studies. The strengths of the fitted Lorentz oscillators are nearly identical to the previously determined values, with \( \epsilon_\infty \approx 4.6 \) (the same value used for the metallic inclusions). Given that the insulating matrix accounts for about 90% of the sample volume, it is not surprising that the vibrational parameters should remain essentially unchanged. Previous Drude-Lorentz fits to the volume-averaged optical conductivity at 35 K yielded \( \omega_{p,D} \approx 430 \) and \( 1/\tau_D \approx 70 \text{ cm}^{-1} \); this result is consistent with the typically low values of \( \omega_{p,D} \) observed in these materials.

If we attribute this average plasma frequency to the fraction \( f \) of the sample that is metallic, then

\[
\frac{\omega_{p,D}}{\omega_{p,D}^{\text{vol}}} = \frac{f}{\sqrt{f}}.
\]

(5)

For \( f \approx 0.11 \), \( \omega_{p,D} = \omega_{p,D}^{\text{vol}} \sqrt{f} \approx 1300 \text{ cm}^{-1} \); this value is almost identical to the fitted EMA value of \( \omega_{p,D} \approx 1320 \text{ cm}^{-1} \). The value for \( \omega_{p,D} \) is still rather small when compared with values of \( \omega_{p,D} \approx 7000–14000 \text{ cm}^{-1} \) observed in other iron-based superconductors. Indeed, for \( \omega_{p,D} \) to rival these values would require a volume fraction of less than 1%. Setting the fraction of metallic inclusions to \( f = 0.005 \) yields the EMA fitted value of \( \omega_{p,D} \approx 6620 \text{ cm}^{-1} \) (close to the value of 6080 cm\(^{-1}\), based on the volume average), and \( 1/\tau_D \approx 38 \text{ cm}^{-1} \); however, the fitted value \( \phi_c \approx 200 \) is quite large, and the overall quality of the fit has decreased significantly. In either case, the temperature dependence of the volume-averaged conductivity was originally described as incoherent at room temperature with a large scattering rate that decreases rapidly with temperature, resulting in a crossover to coherent behavior at low temperatures. However, assuming \( f \approx 0.1 \), the EMA values for \( 1/\tau_D \) suggest that the transport in the metallic regions is always coherent.

The value for \( \phi_c \approx 30 \) yields the rather small value for the depolarization factor \( g_c \approx 0.032 \), which corresponds to an eccentricity \( \epsilon_c \approx 0.93 \) for an oblate spheroid or \( \epsilon_c \approx 0.99 \) in a prolate spheroid; both cases correspond to highly distorted shapes. This condition becomes even more severe for larger values of \( \phi_c \). The layered nature of these materials and the anisotropic transport properties suggest that these distorted shapes overlap or are joined through weak links to form a conducting pathway through the solid, resulting in a predominantly two-dimensional filamentary network or a superconducting aerogel.

In this approach, we have assumed that the highly distorted inclusions overlap to some degree. However, it might also be possible that a large number of inclusions may be isolated, in which case, for spherical particles with \( \epsilon_\infty \approx 1 \) for both the inclusions and the matrix, the effective dielectric function would experience a resonance at \( \omega_0 = \omega_{p,D} \sqrt{(1-f)/f} \approx 720 \text{ cm}^{-1} \) (Maxwell-Garnet theory). However, the values \( \epsilon_\infty \approx 4.6 \) and \( \phi_c \approx 30 \) dramatically lower this resonance \( \omega_0 \approx 100 \text{ cm}^{-1} \). The absence of such a feature in our results suggests that either a continuous distribution of shapes has rendered this feature too broad and too weak to be observed or that there are simply very few isolated inclusions.

B. Energy scales

In the iron-chalcogenide superconductors, the energy scales for the isotropic superconducting energy gaps that are observed in angle-resolved photoemission spectroscopy (ARPES) to open on the hole and electron pockets below \( T_c \) are usually in excellent agreement with the optical gaps observed in the conductivity that develops in the superconducting state. However, these two energy scales appear to be very different in \( K_0.8\text{Fe}_2\gamma\text{Se}_2 \). Although the ARPES estimate of the isotropic optical gap is \( 2\Delta \approx 16–20 \text{ meV} \), the reflectance (and the conductivity) indicates that the energy scale associated with the superconductivity in this material is much smaller \( \approx 8 \text{ meV} \). This difference originates from the inhomogeneous nature of this material. ARPES is insensitive to the insulating matrix and directly probes the formation of a superconducting gap in the metallic (superconducting) inclusions, whereas, the optical properties are a volume-averaging technique, which are sensitive to the Josephson coupling between the superconducting regions. In such a Josephson-coupled system, changes in the reflectance (for instance) will occur not at \( 2\Delta \) but at the renormalized superconducting plasma frequency \( \tilde{\omega}_{p,S} = \omega_{p,S}/\sqrt{\epsilon_{\text{eff}}} \) or at the average value for a distribution of frequencies. Given \( \omega_{p,S} \approx 220 \text{ cm}^{-1} \) and \( \epsilon_{\text{eff}} \approx 18 \) at 50 meV (inset of Fig. 1), then \( \omega_{p,S} \approx 52 \text{ cm}^{-1} \) or \( \approx 6.5 \text{ meV} \), which is very close to the changes in the optical properties observed to occur below \( \approx 8 \text{ meV} \). Thus, due to the inhomogeneous nature of this superconductor, optics and ARPES probe two different quantities \( \tilde{\omega}_{p,S} \) and \( \Delta \), respectively.

C. Parameter scaling

It has been pointed out that a number of the iron-based superconductors fall on the scaling relation initially observed for the cuprate superconductors, where the superfluid density is \( \rho_s \equiv \omega_{p,S}^{\text{vol}} \). In our previous optical paper on \( K_{0.8}\text{Fe}_2\gamma\text{Se}_2 \), the volume-averaged value for the superconducting plasma frequency was determined to be \( \omega_{p,S}^{\text{vol}} \approx 220 \text{ cm}^{-1} \) (Ref. 28). Although this value is quite small, this material does indeed fall on the universal scaling line; however, it does so in a region associated with the response along the \( c \) axis in the cuprates where the superconductivity is due to Josephson coupling between the copper-oxygen planes. From this, it was concluded that the superconductivity was due...
value will represent an average value. As a result, there is some uncertainty attached to the value of $1/\tau_D$. With this caveat in place, the EMA fit to the reflectance just above $T_c$ at 35 K may be used to estimate the dc conductivity of the metallic inclusions $\sigma_{dc} = \omega_{p,S}^2 \tau_D/\rho_{\sigma} \sim 510 \Omega^{-1} \text{cm}^{-1}$. The values for $\omega_{p,S}$ and $\rho_{\sigma}$, once again place this material close to the scaling line, but now the material falls very close to the other iron-chalcogenide superconductors as shown in Fig. 3.

Although $\omega_{p,S}$ is significantly larger than the volume-averaged value, it is still almost an order of magnitude smaller than $\mu SR$ and NMR (Ref. 55) estimates, although adopting a smaller value for the volume fraction $f$ negates this difference. On the other hand, the value of $\lambda_{\text{eff}} \simeq 2.2 \mu m$ in the superconducting regions of $K_{0.3}Fe_{2-x}Se_2$ is in surprisingly good agreement with the in-plane optical estimate of $\lambda \simeq 2 \mu m$ in Rb$_2$Fe$_4$Se$_5$ using an EMA approach.24

FIG. 3. (Color online) The log-log plot of the spectral weight of the superfluid density $N_s = \rho_{0B}/8 \sim \sigma_{dc} T_c$ in the $a-b$ planes for a variety of cuprate superconductors as well as several iron-based superconductors compared with the volume-average and EMA results for $K_{0.3}Fe_{2-x}Se_2$. The dashed line corresponds to the general result for the cuprates $\rho_{0B}/8 \sim 4.4 \sigma_{dc} T_c$, whereas, the dotted line denotes the region of the scaling relation typically associated with Josephson coupling along the $c$ axis. Although the volume-average result signaled a Josephson phase, the EMA result now lies very close to the coherent regime.

to the Josephson coupling of discrete superconducting regions and that the material constituted a Josephson phase.32,51

IV. CONCLUSIONS

The complex optical properties of $K_{0.3}Fe_{2-x}Se_2$ in the normal state have been modeled using the Bruggeman EMA. The volume fraction of the metallic inclusion is estimated to be $f \sim 0.1$; however, the EMA can only be successfully fit to the data if the inclusions are highly distorted, suggesting a filamentary network of conducting regions joined through weak links. The plasma frequency in the metallic inclusions is, therefore, considerably larger than the volume-averaged value $\omega_{p,D} \sim \omega_{p,S}$; however, $\omega_{p,D} \simeq 1320 \text{ cm}^{-1}$ is still much smaller than the values for the plasma frequency observed in other (homogeneous) iron-based superconductors as is the estimate of $\omega_{p,S} \simeq 700 \text{ cm}^{-1}$ (unless volume fractions of less than 1% are considered). The reasonably small values for $1/\tau_D \sim 60-140 \text{ cm}^{-1}$ returned by the EMA fits suggests that the transport in the metallic regions is always coherent and that there is no crossover from incoherent behavior as the temperature is lowered. The inferred $\sigma_{dc} \simeq 510 \Omega^{-1} \text{cm}^{-1}$ just above $T_c$ and the estimated lower bound of $\rho_{\sigma} \sim 4.9 \times 10^2 \Omega \text{cm}^{-2}$ for the metallic (superconducting) inclusions shift this material away from the region on the scaling line associated with Josephson coupling to a region where the majority of (homogeneous) iron-based superconductors is observed to lie.

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