Optical Study of $LaO_{0.9}F_{0.1}FeAs$: Evidence for a Weakly Coupled Superconducting State

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We have studied the reflectance of the recently discovered superconductor $LaO_{0.9}F_{0.1}FeAs$ in a wide energy range from the far infrared to the visible regime. We report on the observation of infrared active phonons, the plasma edge, and possible interband transitions. On the basis of this data and the reported in-plane penetration depth $\lambda_L(0) = 254$ nm [H. Luetkens *et al.*, Phys. Rev. Lett. **101**, 097009 (2008)] a disorder sensitive relatively small value of the total electron-boson coupling constant $\lambda_{\text{tot}} = \lambda_{e\text{-ph}} + \lambda_{e\text{-sp}} \sim 0.6 \pm 0.35$ can be estimated adopting an effective single-band picture.

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The discovery of superconductivity up to 43 K in $LaO_{1-x}F_xFeAs$ [1,2] has established a new family of superconductors with rather high transition temperatures T_c. The substitution of La with other rare earths even yields T_c values above 50 K [3]. In addition to these already fascinating reports, both experimental and theoretical studies consider possible unconventional multiband behavior [4,5]. Various scenarios for the superconducting mechanism have been proposed or excluded [6–12]. A more general related problem under debate [5,13–19] being of considerable interest is the strength of the correlation effects in these doped oxypnictides governed by the local Coulomb interactions U_d and J_d at Fe sites.

The crystals of $LaO_{1-x}F_xFeAs$ are formed by alternating stacking of $LaO_{1-x}F_x$ and FeAs layers. The electronic structure of these new systems has been mainly addressed by theoretical studies so far. They predict that LaOFeAs and the related compounds harbor quasi two-dimensional electronic bands as is suggested by the layered crystal structure already [4–9]. The LDA (local density approximation) Fermi surface of the doped compound consists of four cylinderlike sheets . Hence, the electronic behavior is expected to be highly anisotropic concerning, e.g., charge transport and superconducting properties such as the penetration depth $\lambda_L(T)$ and the upper critical magnetic field. In addition, calculations have provided the expected phonon density of states (PDOS) which might be relevant for the symmetry of the superconducting order parameter even in the case of a dominant magnetic coupling in spite of the predicted weak electron-phonon (e-ph) coupling [6,20]. So far, optical data are available for the far-infrared region only, where indications for the opening of a gap in the superconducting state have been found, and infrared active phonons can be observed [21]. To the best of our knowledge a phenomenological estimate of the electron-boson (e-b) coupling strengths is still missing and the present work provides a first step in that direction.

Here we report on a study of the optical properties of $LaO_{0.9}F_{0.1}FeAs$ via reflectance measurements. We have determined the reflectance of powder samples in the energy range from 0.009 up to 3 eV at $T = 300$ K. Our data reveal two prominent and two weaker infrared active phonon modes, and provide evidence for the in-plane plasma energy at ≈ 0.4 eV as well as the appearance of electronic interband transitions at higher energies. These data are used to estimate the strength of the total e-b coupling λ_{tot} responsible for the ''mass enhancement'' in the plasma energy of the paired electrons without specifying the nature of this coupling. We compare its value with various microscopic estimates in the literature based on spin fluctuations and electron-phonon contributions as well as the empirical unscreened plasma energy with uncorrelated L(S)DA (local spin density approximation) predictions.

A polycrystalline sample of $LaO_{0.9}F_{0.1}FeAs$ was prepared as described in our previous work [22] and in Ref. [23]. For the optical measurements reported here the pellets have been polished to obtain appropriate surfaces. In order to characterize the superconducting properties, zero field cooled (shielding signal) and field cooled (Meissner signal) magnetic susceptibility in external fields $H = 10$ Oe ... 50 kOe have been measured using a SQUID magnetometer. The resistivity has been measured with a standard 4-point geometry employing an alternating dc current. A value of $T_c \approx 26.8$ K has been extracted from these data as shown in Fig. 1.

The reflectance measurements have been performed using a combination of Bruker IFS113v/IFS88 spectrometers. This allows us to determine the reflectivity in an energy range from 0.009 up to 3 eV. The measurements have been carried out with different spectral resolutions depending on the energy range; these vary form 0.06 to 2 meV. Since the observed spectral features are significantly broader than these values, this slight variation in resolution does not impact our data analysis. All reflectance measurements were performed at $T = 300$ K and a pressure of 4 mbar.

In Fig. 2 we show the reflectance of our $LaO_{0.9}F_{0.1}FeAs$ sample (note the logarithmic and linear energy scales in the two panels, respectively). These data reveal a number of spectral features with different origin. At low energies (see

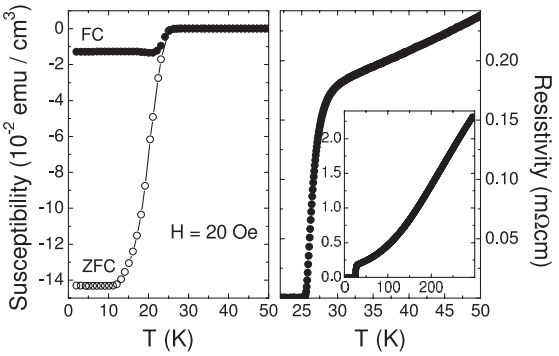

FIG. 1. Left: field cooled (FC) and zero field cooled (ZFC) magnetic susceptibility for the $LaO_{0.9}F_{0.1}FeAs$ samples studied here. Right: T dependence of the resistivity near T_c.

left panel of Fig. 2) there are two prominent structures at about 12 and 55 meV and two weaker features in between near 25 and 31 meV, which most likely stem from the excitation of infrared active phonons. We attribute the two strong features to phonons that are polarized along the **c** axis of the lattice, since the dielectric screening in this direction is expected to be much weaker as a result of the rather two-dimensional electronic system. An assignment of the two weaker phonons is not possible at the present stage, and future work on single crystals will help to clarify their polarization character. A comparison with a calculated PDOS [6] allows an assignment of the higher energy phonon (55 meV) to oxygen modes, since above about 40 meV only these vibrations can be expected. In addition, the calculated PDOS has a maximum near 12 meV [6] with contribution from all elements in the structure, which would correspond to the strong lowest energy feature in Fig. 2, and the calculations also predict two further maxima which could be associated with weaker structures in our reflectance spectrum.

At higher energies the reflectance significantly drops between 0.13 and 0.4 eV; thereafter it shows a small

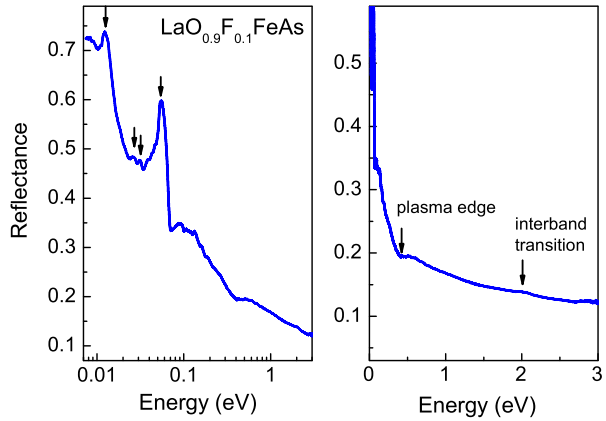

FIG. 2 (color online). Reflectance of a polished sample. The energy is given in a logarithmic (left) and linear (right) scale. The arrows in the left highlight the low-energy excitation features ascribed to phonons. In the right the arrows denote the plasma edge and the 2 eV interband transition.

increase until about 0.6 eV before it starts to slightly decrease again. In addition, at about 2 eV another small upturn is visible. We attribute the two small features at about 0.6 and 2 eV to a weak electronic interband transition at the corresponding energies. This observation is in accord with the behavior of the optical conductivity $\sigma(\omega)$ predicted by Haule *et al.* using dynamical mean-field theory (DMFT), where As $4p$ to Fe $3d$ interband transition (IB_{pd}) slightly below 2 eV and a weak feature near 0.6 eV for the undoped parent compound LaOFeAs have been found. The steepest edge centered at about 250 meV with an high-energy onset at about 395(20) meV represents the plasma edge or plasma energy of $LaO_{0.9}F_{0.1}FeAs$, which is observed at a relatively low value. In consideration of the quasi-2D character of the electronic states in $LaO_{0.9}F_{0.1}FeAs$ and the expected strong anisotropy of the plasma energy for light polarized ∥ to the (**a**, **b**) and **c** crystal axes, respectively, the onset of the plasma edge (PE) in Fig. 2 gives a reasonable value of the in-plane value [i.e., for light polarized within the (**a**, **b**) crystal plane] [24].

Within a simple Drude model, this value allows a first, crude estimate of the charge carrier density in $LaO_{0.9}F_{0.1}FeAs$. Within this model, the plasma energy, ω_p, is given by $\omega_p^2 = ne^2/m\varepsilon_0\varepsilon_\infty$, with n, e, m, and ε_∞ representing the charge carrier density, the elementary charge, the effective mass of the charge carriers, and the background dielectric screening due to higher lying electronic excitations [25]. Taking the effective mass equal to the free electron mass, and $\varepsilon_\infty \sim 10$–$12$ [26], one would arrive at a rather low charge density of about 4×10^{-20} cm^{-3}. We note that this estimate ignores the strongly anisotropic nature of the electronic structure near the Fermi energy E_F. Our LDA-FPLO [full potential localized orbital minimum-basis code (version 7)] [27] band structure calculations for the nonmagnetic ground state provide a value for the plasma frequency within the (**a**, **b**) crystal plane of $\Omega_p^{\mathrm{LDA}} = 2.1$ eV and a small value for a polarization along the **c** axis of 0.34 eV in accord with 2.3 eV and 0.32 eV given in Ref. [6]. Note that these "bare" values do not describe the screening through ε_∞ caused by interband transitions. In order to get the measured $\omega_p = 0.395$ eV an unusually large value of $\varepsilon_\infty \sim 28.3$ to 33.9 would be required, which seems to be unrealistic [26], and instead $\varepsilon_\infty \sim 12$ would be expected. Then, for the empirical unscreened plasma energy Ω_p a value about 1.37 eV is expected on the basis of our data. For a deeper insight we calculated at first the interband contribution to the dielectric function from the in-plane DMFT optical conductivity between 1 and 6 eV for the undoped LaOFeAs given in Fig. 5 of Ref. [13]. From its static value we obtained $\varepsilon_\infty = 5.4$, only. Note that generally smaller values for the onsite Coulomb repulsion U_d would result in larger ε_∞ values due to an energy shift of the interband transition from the lower Hubbard band and the less suppressed combined density of states for the IB_{pd} transitions which vanishes at the metal-insulator transition expected at a critical U_d slightly above

4 eV [13]. Hence, a corresponding systematic DMFT study with lower U_d and J_d values would be of interest. In contrast to Haule *et al.* [13], Shorikov *et al.* [14] argue that LaOFeAs is in an intermediate U_d regime but strongly affected by the value of the Hund's rule exchange J_d. Hence, ε_∞ should significantly increase and the estimate given above can be regarded as a *lower* bound. But it provides an unrealistic estimate for ε_∞ itself. The expected U_d dependence of ε_∞ is shown schematically in Fig. 3. It points to strongly reduced U_d values in accord with the suggestions of Refs. [14,19]. Thus, ε_∞ provides a convenient direct measure for the strength of the correlation regime, even for small U_d values above 2 eV.

In addition, there is a direct relation between the plasma energy as measured with optical techniques and the London penetration depth in the superconducting state. Within a BCS approach, these two parameters are inversely proportional to each other. Recently, the in-plane penetration depth, $\lambda_{a,b}$, for $LaO_{0.9}F_{0.1}FeAs$ has been determined using muon spin relaxation to be $\lambda_L^{a,b}(0) = 254(2)$ nm [22,28]. This value slightly exceeds those found for optimally doped high-T_c cuprates (HTC). This suggests that the plasma energy of $LaO_{0.9}F_{0.1}FeAs$ should be smaller than that of HTC. Their ω_p is found near 1 eV [29–32] for optimal doping (i.e., the highest T_c), and taking into account the different penetration depths $\lambda_L^{a,b}$ one would expect a plasma energy in $LaO_{0.9}F_{0.1}FeAs$ that is close to the measured value. Here we take into account that λ_L is renormalized by the mass enhancement factor as a consequence of the always present *e-b* coupling measured by the Fermi surface averaged coupling constant λ_{tot}, whereas the plasma energy is not. This asymmetry in the renormalization is based on very different temperatures and energies probed in both measurement: nearly $T = 0$ and low ω for the penetration depth vs high energies and high T for the PE; see also Fig. 7 in Ref. [33] and the *Note added*. In other words, we employ both the $T = 0$ and the asymptotic high-energy limits of the coupling constant being in general ω and T dependent. In the effective single-band approach this relation can be rewritten in convenient units as [34]

$$\alpha\Omega_p[\text{eV}] = \sqrt{[n/n_s(0)][1 + \lambda_{\text{tot}}(0)](1 + \delta)}, \qquad (1)$$

where $\alpha = \lambda_L(0)$ [nm]/(197.3 nm) is the experimental (**a**, **b**)-plane penetration depth extrapolated to $T = 0$, $\Omega_p = \sqrt{\varepsilon_\infty}\omega_p$ denotes the empirical unscreened plasma energy, $n_s(0)$ is the density of electrons in the condensate at $T = 0$ and n the total electron density which contributes to the unscreened Ω_p, $\delta = 0.7\gamma_{\text{imp}}/[2\Delta(0)]$ is the disorder parameter which vanishes in the clean limit. Notice that all three factors under the root are ≥ 1. In the quasiclean limit $\delta < 1$, for $n = n_s(0)$, and using $\varepsilon_\infty = 12$ [26], $\delta = 0.93$ K for $\gamma_{\text{imp}} = 125$ K estimated from resistivity data [35] and $\lambda_L(0) = 254$ nm [22,28] one estimates $\lambda_{\text{tot}} \sim 0.61$; i.e., the superconductivity is in a weak coupling regime for our samples with $T_c = 27$ K. The effect of the quantity $\varepsilon^* = \varepsilon_\infty/(1 + \delta)$ on the coupling strength is shown in Fig. 3. Note that a substantial impurity scattering ($\delta > 1$) beyond our moderate disorder regime should further reduce λ_{tot}. We note once more that our empirical $\Omega_p \approx 1.37$ eV differs clearly from the LDA prediction of 2.1 eV for the nonmagnetic ground state and slightly from 1.6 eV for the rigid band estimate of the antiferromagnetic state within the LSDA.

The microscopic nature of the empirically estimated total *e-b* coupling constant and especially its decomposition into the interaction with various boson modes remains unclear. Since the experimental optical mass deviates by a factor of 2.35 from the LDA predictions based on the neglect of antiferromagnetic fluctuations and correlation effects (and the LSDA by a factor of 1.37) it is also unclear to what extent the calculated weak *e*-ph coupling $\lambda_{e\text{-ph}}^{\text{LDA}} \leq 0.21$ [6] can be trusted. These deviations might be caused by the filling of the two (three) Fermi surface hole pockets [13] as a result of correlations or a hidden spin-density wave [17], or another many-body effect. Adopting nevertheless such a weak value for $\lambda_{e\text{-ph}}$ one is left with a nonphonon contribution of 0.2 to 0.4. Since the screening by the interband transitions is affected by the value of U_d, Fig. 3 suggests the stronger the correlation the weaker is the empirical *e-b* coupling strength.

In general, a classical *e*-ph mechanism seems to be ruled out by the weakness of λ_{ph} and the smallness of typical phonon energies except an exotic situation with an attractive Coulomb interaction $\mu^* < 0$ due to a *negative* dielectric response at large wave vectors [36] or huge pnictide polarizabilities [37]. Multiband effects ignored here might lead to somewhat larger λ values since the low-T penetration depth and $\lambda_L(0)$ in particular, might be affected by the weakest coupled group of electrons with the smallest gaps and the largest Fermi velocities (Ω_p) whereas the region near T_c is dominated by particles with the largest gaps [38]. The corresponding estimates, e.g., within a two-band

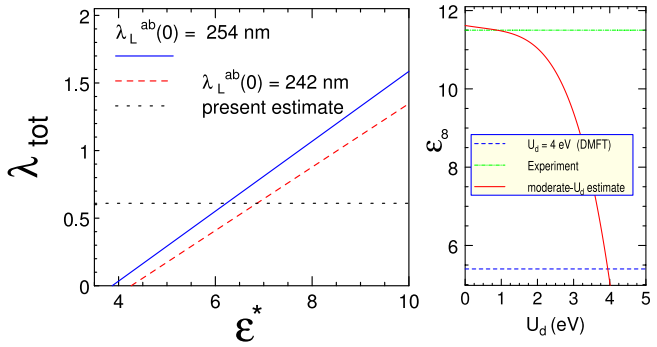

FIG. 3 (color online). Empirical relation between the total coupling constant from the mass enhancement entering the penetration depth vs interband screening modeled by the disorder renormalized dielectric background constant $\varepsilon^* = \varepsilon_\infty n_s/n(1 + \delta)$. The $\lambda_L^{ab}(0)$ values from Refs. [22,28] (left). The improved screening measured by the background dielectric constant ε_∞ vs reduced on-site repulsion U_d on Fe sites (right).

model are postponed to the time when more data will be available.

To summarize, we have studied the optical response of $LaO_{0.9}F_{0.1}FeAs$ powder samples in a wide energy range. Our reflectance data reveal four features near 12, 25, 31, and 55 meV we assigned to phonon excitations. Furthermore, our data allow us to extract the screened in-plane plasma energy to about 0.4 eV and two interband excitations near 0.6 and 2 eV. The subsequent analysis of the reported penetration depth suggests a quasiclean limit, weak coupling superconducting regime. The analysis of ε_∞ in the correlated regime at $U_d = 4$ eV points to a substantially reduced Coulomb repulsion. This is in accord with Refs. [14,19,37]. Further systematic studies including various dopings, oriented films, single crystals, and other Fe based superconductors are desirable.

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Note added.—Preparing an amended manuscript we learned about an ellipsometry study of $LaO_{0.9}F_{0.1}FeAs$, where $\tilde{\Omega}_p = 0.61$ eV has been reported applying the effective medium approximation (EMA) to obtain $\sigma_{\text{eff}}(\omega)$ for a polycrystalline sample [26]. However, for spherical grains and $\sigma_{ab} \gg \sigma_c$ as expected for a quasi-2D metal (except for the unscreened c-axis polarized ir phonon related peaks), EMA predicts $\sigma_{\text{eff}} \approx 0.5\sigma_{ab}$ which corresponds to $\tilde{\Omega}_p^{ab} = 0.86$ eV Since $\tilde{\Omega}_p$ has been derived mainly from the low-ω, damped Drude region (below 25 meV) it is still renormalized by the e-b coupling, where a high-energy boson well above 25 meV has been assumed to explain the high T_c at weak coupling. Using our $\lambda = 0.61$ one estimates $\Omega_p = 1.23$ eV, close to 1.37 eV suggested above [39]. Finally, the analysis of the Pauli limiting (PL) behavior for the closely related $LaO_{0.9}F_{0.1}FeAs_{1-\delta}$ system yields a similar value $\lambda \approx 0.6$ to 0.7 as derived from the strong coupling correction for its PL field $B_P(0) = 102$ T [40]. Rather similar phonon-peaks as reported here, have been detected recently also for $SmO_{1-x}F_xFeAs$ [41].

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