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Multiband conductivity and a multigap superconducting phase in V_3Si films from optical measurements at terahertz frequencies

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The possibility of multiband conductivity and multigap superconductivity is explored in oriented V_3Si thin films by means of reflectance and transmittance measurements at terahertz frequencies. The temperature dependence of the transmittance spectra in the normal state gives evidence of two bands contributing to the film conductivity. This outcome is consistent with electronic structure calculations performed within density functional theory. On this basis, we performed a detailed data analysis and found that all optical data can be consistently accounted for within a two-band framework, with the presence of two optical gaps in the superconducting state corresponding to $2\Delta/kT_c$ values close to 1.8 and 3.8.

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Large interest has been devoted[1,2] to multiband superconductivity after the discovery of two bands with two distinct superconducting gaps in MgB_2.[3] Very recently, the interest in multiband superconductivity has been renewed by the discovery of the Fe-As based superconductors, where the presence of multiple bands is well established.[4] It is worth noting that two-gap superconductivity has been considered theoretically since the fifties,[5] and experimentally observed in transition metals,[6] in the A15 compound Nb_3Sn (Refs. 7 and 8) and in $MgCNi_3$.[9] However, only after the discovery of superconductivity in MgB_2, clear cut evidence of two-gap superconductivity and of its implications was obtained, since in this compound the multiband, multigap character is emphasized by the exceptionally low interband scattering.[10]

In the case of the A15 V_3Si system, a spread in the gap values corresponding to $2\Delta/kT_c$ extending from 1.0 to 3.8 has been reported.[11,12] Recently, the electrodynamic response in the microwave region gave evidence of two gaps,[13] while muon spin rotation measurements were consistent with a single-gap model.[14] It is also worth noting that V_3Si has been recently treated as a two-band (2-b) system,[15] but existing band structure calculations[16,17] do not provide detailed enough information.

In this contradictory scenario, we investigate in the present work the possible two-band, two-gap character of V_3Si, by performing both an infrared spectroscopy study and electronic structure calculations within density functional theory (DFT).

Infrared (IR) spectroscopy is a powerful tool to study the properties of a conducting system. In the normal state (N state), the Drude model for the frequency-dependent complex conductivity $\tilde{\sigma}_N=\sigma_{1N}+i\sigma_{2N}$ can be employed to describe the optical response of free-charge carriers.[18] As shown in the MgB_2 case,[19,20] it is thus possible to determine the contributions of different i bands and thus the corresponding plasma frequencies Ω_i and scattering rates γ_i. In the superconducting state (S state), on the basis of the Bardeen-Cooper-Schrieffer model for the complex conductivity $\tilde{\sigma}_S=\sigma_{1S}+i\sigma_{2S}$,[21] far-IR/terahertz measurements can be of particular importance since a mark of the superconducting gap Δ can be observed at $\hbar\omega\sim 2\Delta$ (optical gap) for an isotropic s-wave Bardeen-Cooper-Schrieffer superconductor. In particular, a maximum at the optical gap is expected either in the ratio R_S/R_N (for a bulk sample) or in the ratio $\mathcal{T}_S/\mathcal{T}_N$ (for a thin film), where $R_S(\mathcal{T}_S)$ and $R_N(\mathcal{T}_N)$ are the frequency-dependent reflectances (transmittances) in the S and N state, respectively.[21] In the MgB_2 case,[19] evidence of the two gaps was detected in the R_S/R_N spectrum of an ultraclean film[20] by assuming a parallel sum of the conductivity of two independent bands and by using the model $\tilde{\sigma}_S$ introduced by Zimmermann *et al.*[22] which generalizes the Bardeen-Cooper-Schrieffer one to arbitrary T and γ values (Zimmermann model).

We performed IR measurements on high-quality V_3Si textured films. Details on the film growth by pulsed laser deposition and on their properties are reported elsewhere.[23] We studied two films grown on $LaAlO_3$ (LAO) (001) 0.5 mm thick substrates, which exhibit preferential (210) orientation along the out-of-plane direction. The first film of thickness $d=180$ nm (film d180) has good transport properties (resistivity at 300 K close to 200 $\mu\Omega$ cm, residual resistivity ratio $RRR=8$) and $T_c=16.1$ K; the second film, 33 nm thick (film d033), has worst transport properties ($RRR=4.5$) and a slightly lower T_c value ($T_c=15.3$ K).

We first performed measurements of the IR reflectance $R(\omega)$ of film d180 as a function of temperature (not shown).

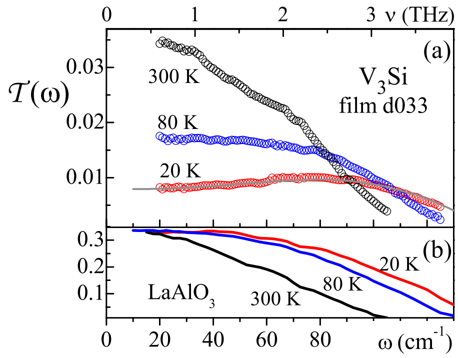

FIG. 1. (Color online) (a) Transmittance spectra $\mathcal{T}(\omega)$ of film d033 at selected temperatures for $T > T_c$ in the THz region. The 20 K spectrum is compared with the two-band (2-b) best fit. (b) $\mathcal{T}(\omega)$ spectra of a $LaAlO_3$ substrate (0.5 mm thick) at the same temperatures from Ref. 24.

The $R(\omega)$ spectrum at 300 K is in very good agreement with the result of previous measurements.[17] We attempted an analysis of the measured spectra by using the standard Drude-Lorentz model. Unambiguous results were not obtained since at any temperature a very broad Drude contribution[17] is partially overwhelmed by intense lorentzian contributions associated to the interband transitions occurring at rather low frequencies because of the complex electronic structure of V_3Si.[16,17]

We then measured the transmittance $\mathcal{T}(\omega)$ spectrum of film d033. Measurements were performed between 300 and 20 K in the terahertz (THz) region (here defined by photon energies below 16 meV, frequency $\omega < 130$ cm^{-1} or $\nu < 4$ THz), where the LAO substrate is partially transparent. $\mathcal{T}(\omega)$ spectra of film d033 are reported in Fig. 1(a) at selected temperatures, those of LAO (Ref. 24) at the same temperatures in Fig. 1(b) for comparison. The spectral shape of the 300 K $\mathcal{T}(\omega)$ spectrum is qualitatively similar to that of LAO; the transmitted intensity is strongly reduced by the above discussed broad Drude contribution in the film conductivity. On the contrary, the spectral shape of the 20 K $\mathcal{T}(\omega)$ shows a broad maximum and then decreases with decreasing frequency. This result can be qualitatively explained by the presence of a second sharp Drude contribution, which narrows with decreasing temperature thus reducing the transmitted intensity at very low frequencies.

In order to better understand this result, we carried out electronic structure calculations within DFT on V_3Si. We calculated the Fermi velocities starting from the energy bands shown in the inset of Fig. 2. We used the experimental lattice constants a and c,[25] and calculated the dimerization of V chains along x and y directions to be $0.0025a$. The Fermi level E_F of V_3Si is crossed by four bands (two bands with similar character and small density of states are grouped together) with mostly V $3d$ character; with the chain direction along z, orbitals $d_{x^2-y^2}$ and d_{z^2} on one side and d_{xz} and d_{yz} on the other produce narrow and wide bands, respectively.

In Fig. 2, we plot the distribution of values of the moduli of Fermi velocities[26] for the bands crossing E_F; the distributions are normalized to the densities of states of the same bands, so that the average squared velocity times the integral

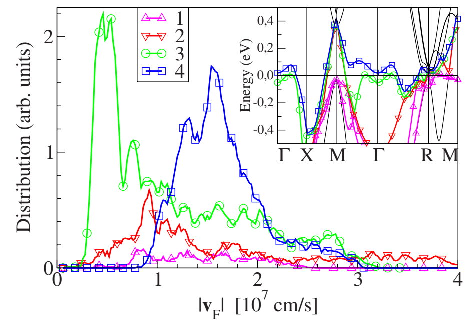

FIG. 2. (Color online) Distribution of moduli of the Fermi velocities for the bands crossing E_F. Inset: energy bands along the symmetry lines of the Brillouin zone; the different symbols identify the bands as in the inset.

of the distribution provides, apart from numerical factors, the contribution to the square of the total plasma frequency $\Omega_{tot} = 3.34$ eV. While it is difficult to assign a definite orbital character to each band throughout the whole Brillouin zone, we can see that the band separation naturally provides different average velocities, showing an overall bimodal distribution given by the bands n. 3 and 4, while the remaining bands provide a smaller contribution to plasma frequencies due to their smaller density of states. The bimodal distribution resulting from our calculations indicates that the V_3Si conductivity in the N state can be safely described in terms of two bands characterized by different Ω values (two-band, 2-b model). It remains to be verified whether the presence of the two bands reflects in the opening of two distinct superconducting gaps, as in the MgB_2 case.

To this aim we measured at $T \leq 20$ K the $R_S(T)/R_N$ [where $R_N = R(20\ \mathrm{K})$] spectra of the d180 film [see Fig. 3(a)] and the $\mathcal{T}_S(T)/\mathcal{T}_N$ [where $\mathcal{T}_N = \mathcal{T}(20\ \mathrm{K})$] spectra of the d033 film [see Fig. 4(a)]. These measurements were made by cycling the temperature in the 6–20 K range, without collecting reference spectra. In this way one avoids any variation in the sample position and orientation, which may yield frequency-

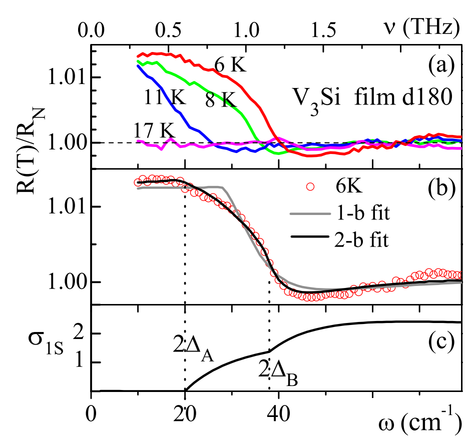

FIG. 3. (Color online) (a) $R(T)/R_N$ spectra of film d180 at selected temperatures in the Thz region. (b) R_S/R_N (with $R_S = R(6\ K)$) spectrum compared with the two-band (2-b) and one-band (1-b) best fits. In the 1-b case, best-fit parameters are $\Omega = 2.35$ eV, $\gamma = 125$ cm^{-1}, and $\Delta = 14$ cm^{-1}. (c) σ_{1S} (in units $10^4\ \Omega^{-1}$ cm^{-1}) of V_3Si from 2-b model.

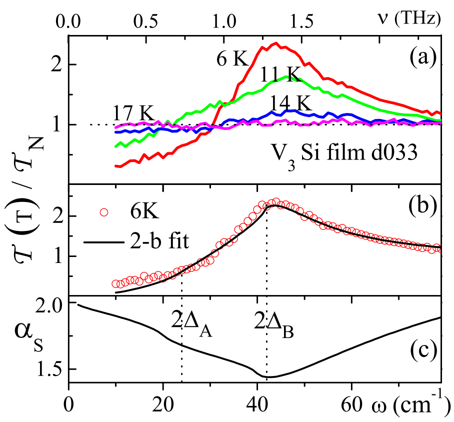

FIG. 4. (Color online) (a) $\mathcal{T}(T)/\mathcal{T}_N$ spectra [with $\mathcal{T}_N=\mathcal{T}(20\text{ K})$] of film d033 at selected temperatures in the THz region. (b) $\mathcal{T}_S/\mathcal{T}_N$ spectrum compared with the two-band (2-b) best fit ($\Omega_A=3.35$ eV, $\gamma_A=1000$ cm^{-1}, $\Delta_A=12$ cm^{-1}, $\Omega_B=1.24$ eV, $\gamma_B=75$ cm^{-1}, $\Delta_B=21$ cm^{-1}). (c) absorption coefficient α_S (in units 10^4 cm^{-1}) of V_3Si from 2-b model.

dependent systematic errors in $R(\omega)$ and $\mathcal{T}(\omega)$. All measurements in the THz region were made by employing synchrotron radiation at the infrared beamline SISSI (Ref. 27) at the synchrotron Elettra (Trieste, Italy) and at the BESSY storage ring (Berlin, Germany), where coherent synchrotron radiation is available. We remark that a high-flux synchrotron source allows a high accuracy in the detection of small effects in the $R(T)/R_N$ spectra, and overcomes the problem of the very low intensity transmitted by the film+substrate system. We also note that $\mathcal{T}_S/\mathcal{T}_N$ [where $\mathcal{T}_S=\mathcal{T}(6\text{ K})$] is affected by the superconducting transition much more than R_S/R_N [where $R_S=R(6\text{ K})$]. Indeed the transmitted radiation probes the entire film thickness and is thus dominated by the effect of the absorptive processes, which significantly increase below T_c when the pair-breaking mechanism originates.[21,28]

We first notice that the $R(T)/R_N$ spectrum increases on decreasing temperature [see Fig. 3(a)], until R_S/R_N reaches a maximum and becomes nearly constant below 20 cm^{-1}. This indicates the presence of a superconducting gap Δ close to 10 cm^{-1}. Indeed, for $\omega\rightarrow 0$, the reflectance R_N of a conducting system tends to 1 for a bulk system, to a slightly lower value for a thin film. Therefore, since R_S approaches 1 at $\omega=2\Delta$, R_S/R_N exhibits a maximum around 2Δ in the case of a bulk sample, remains nearly constant below 2Δ in the film case. As to the $\mathcal{T}(T)/\mathcal{T}_N$ data [see Fig. 4(a)], a maximum develops on decreasing T until the $\mathcal{T}_S/\mathcal{T}_N$ exhibits a well defined peak around 40 cm^{-1}, which indicates the presence of a superconducting gap Δ around 20 cm^{-1}.

For a detailed analysis of the measured spectra based on the 2-b model, the conductivity of V_3Si can be described by the sum of two Drude terms in the N state ($\tilde{\sigma}_N$, with parameters Ω_i, γ_i, $i=A,B$), and by the sum of two Zimmermann terms in the S state, as discussed above ($\tilde{\sigma}_S$, with parameters T, Ω_i, γ_i, Δ_i, $i=A,B$). From $\tilde{\sigma}$, by using standard relations,[18] it is possible to compute the model refractive index $\tilde{n}=n+ik$ of V_3Si in both the N and S state. The transmittance and reflectance spectra of the film+substrate system in both states can then be evaluated by means of an exact procedure[29] which requires thickness and n and k values for both film and substrate. For LAO, n and k in the THz region were obtained from previous measurements of transmittance and reflectance of a LAO substrate[24] through a numerical, model independent procedure.[30]

We first analyzed the R_S/R_N spectrum on the basis of the 2-b model, by performing a 6 parameter fit (Ω_i, γ_i, Δ_i, $i=A,B$). The 2-b best-fit curve does well describe experimental data, while unsatisfactory results are obtained by considering one band only [one-band (1-b), 1-b model], as shown in Fig. 3(b). The 2-b best fit parameter values are $\Omega_A=3.7\pm0.1$ eV, $\Omega_B=1.00\pm0.05$ eV, $\gamma_A=800\pm40$ cm^{-1}, $\gamma_B=45\pm4$ cm^{-1}, $\Delta_A=10\pm1$ cm^{-1}, and $\Delta_B=19\pm2$ cm^{-1}, where the quoted uncertainties represent the range over which each parameter can be varied without degrading appreciably the fit quality. Note that, in the 2-b approach, the A-band contribution has a crucial effect, as it gives a vanishing σ_{1S} only below $2\Delta_A$ [see Fig. 3(c)]. Since, in general, R_S approaches 1 when σ_{1S} vanishes, this explains why only the Δ_A gap is well evident in the R_S/R_N spectrum.

In order to verify the compatibility of this fit with first principle results, we calculated plasma frequencies from our Fermi velocities. In particular, we integrated over the Brillouin zone separating the low and high $\mathbf{v}_F$ regions (setting an arbitrary but plausible cutoff at 10^7 cm/s; see Fig. 2). While this procedure is only qualitative, it may be justified by the presence of clearly separable ranges of $\mathbf{v}_F$, in turn deriving from different orbital natures. We obtain $\Omega_A=3.21$ eV and $\Omega_B=0.81$ eV. In comparing these results with those of the fitting procedure, it is worth noting that the best-fit values of Ω_A and Ω_B, and thus of Ω_{tot}, can be overestimated because the interband transitions discussed above may give non negligible contributions to $\tilde{\sigma}_N$ even at very low frequencies. This effect is minimized in the best-fit Ω_A/Ω_B ratio (3.7 ± 0.3), which results to be in a remarkably good agreement with the computed one (3.96).

In analyzing the transmittance spectra of film d033, we verified that only the 2-b model well describes $\mathcal{T}(\omega)$ at all temperatures, but the resulting Ω_i and γ_i values are not unambiguously determined. We thus used a procedure which simultaneously fits both $\mathcal{T}_N$ and $\mathcal{T}_S/\mathcal{T}_N$, thus imposing important constraints in the fitting procedure. Good fits of both $\mathcal{T}_N$ [see Fig. 1(b)] and $\mathcal{T}_S/\mathcal{T}_N$ [see Fig. 4(b)] were obtained, with Ω_i values slightly lower, γ_i values slightly higher than those found in fitting the R_S/R_N spectrum. This can be simply explained by the worse conducting properties of film d033 with respect to film d180. As to the gap values, nearly equivalent fits are obtained for Δ_A ranging from 11 to 16 cm^{-1}, while $\Delta_B=21.0\pm0.5$ cm^{-1} is well determined since it corresponds to the peak in $\mathcal{T}_S/\mathcal{T}_N$. The transmission measurement, dominated by the absorption process, thus permits to unambiguously establish the Δ_B value which was more poorly defined in the R_S/R_N measurement. The higher sensitivity of $\mathcal{T}_S/\mathcal{T}_N$ to the Δ_B gap is a consequence of the frequency dependence of the absorption coefficient α_S of V_3Si in the S state. Indeed, α_S, as evaluated with standard relations[18] through the 2-b model, only exhibits a well defined minimum around $2\Delta_B$ [see Fig. 4(c)].

In summary, we addressed the debated problem of multiband, multigap nature of V_3Si by means of reflectance and transmittance measurements in the THz region on high-quality oriented films. Experimental results indicate the presence of two bands contributing to the V_3Si conductivity in the normal state, and of two optical gaps in the superconducting state. Electronic structure calculations within density functional theory showed that the distribution of the modulus of the Fermi velocity exhibits a clear bimodal character, which indicates that the V_3Si conductivity in the normal state can be safely described in terms of two bands, characterized by different plasma frequencies. On this basis, we performed a detailed data analysis and found that all optical data can be consistently accounted for within a two-band framework, with the presence of two optical gaps corresponding to $2\Delta/kT_c$ values close to 1.8 and 3.8.

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