Physica C 235-240 (1994) 1085-1086
North-Holland

PHYSICA C

The use of far-infrared ellipsometry in the study of high-temperature superconductors: possibilities and limitations

K. Kamarás[*], D. van der Marel[a], C. C. Homes and T. Timusk[b]

[a] University of Groningen, Nijenborgh 4, NL 9747 AG Groningen, The Netherlands

[b] Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, Canada L8S 4M1

We examine the possibilities of applying spectroscopic ellipsometry for the determination of the far-infrared dielectric function of high temperature superconductors. On the example of $YBa_2Cu_3O_7$ at 20 K we show that the potential of the method lies in the accurate determination of ϵ_1 rather than the details of the onset of ϵ_2.

Spectroscopic ellipsometry, highly succesful in determining the dielectric function of high temperature superconductors in the visible and ultraviolet, has been recently extended towards lower frequencies and proven to give reliable results on $YBa_2Cu_3O_{7-\delta}$ [1,2]. In this paper, we want to take a realistic look at what information the method can yield regarding the open questions in the infrared region. $YBa_2Cu_3O_{7-\delta}$ is particularly well suited for this purpose, as its in-plane dielectric function is well known and verified by different techniques [1-3] and c-axis data at low temperature have recently been obtained [4]. We have used these experimental dielectric functions as input into the standard expression for a uniaxial medium [5] to calculate various quantities relevant to an ellipsometry measurement.

Ellipsometry measures the (complex) reflectivity ratio of *s*- and *p*-polarized light incident on the sample surface at an oblique angle:

$$\tilde{\rho} = \tilde{r}_p/\tilde{r}_s = \tan\psi e^{-i\Delta} \qquad (1)$$

Two quantities per frequency are obtained, usually expressed in the form of the ellipsometric angles ψ and Δ. From these, ϵ_1 and ϵ_2, respectively, can be determined through analytical expressions *if and only if* the isotropic two-phase model (i.e. assuming the sample being a semi-infinite, isotropic medium) [5] is adequate. In other cases (anisotropic or multilayer samples) additional information is required and numerical methods are used for evaluation. Such additional information can be obtained in principle by measuring at more than one angle of incidence. In the case of $YBa_2Cu_3O_{7-\delta}$ this is hardly a possibility, as illustrated in Fig 1, showing the dependence of the ellipsometric angles at 300 cm^{-1} on the angle of incidence. For the phase information to be meaningful, Δ has to be significantly different from 180°; this occurs in a very narrow range above $\theta = 80°$, where the curve is very steep. Our calculations, in agreement with experiment [1,2] show, however, that the dominant *ab*-plane response is largely unaffected by the c-axis structure. This insensitivity on the c-axis is not necessarily a universal feature of all high T_c superconductors, since in $Tl_2Ba_2Ca_2Cu_3O_10$ the c-axis phonons have been observed in reflectivity at 45° [6].

A serious shortcoming of reflectivity measurements has been addressed repeatedly in the past: the inevitable scaling errors in R (due to geometrical limitations when measuring against a reference) lead to large fluctuations in ϵ_2 (and consequently in the optical conductivity) when R approaches unity. This effect obscures the onset of absorption at low frequency. We examined the question whether the error can be reduced in ellipsometry and show our results in Fig. 2. The solid lines are calculated from the *ab*-plane reflectivity of Ref. [3], assumed to be 1 below 130

---

[*] Permanent address: Research Institute for Solid State Physics, Hungarian Academy of Sciences, P.O.Box 49, H 1525 Budapest, Hungary. A. von Humboldt Foundation Fellow 1992-93.

SSDI 0921-4534(94)01106-0

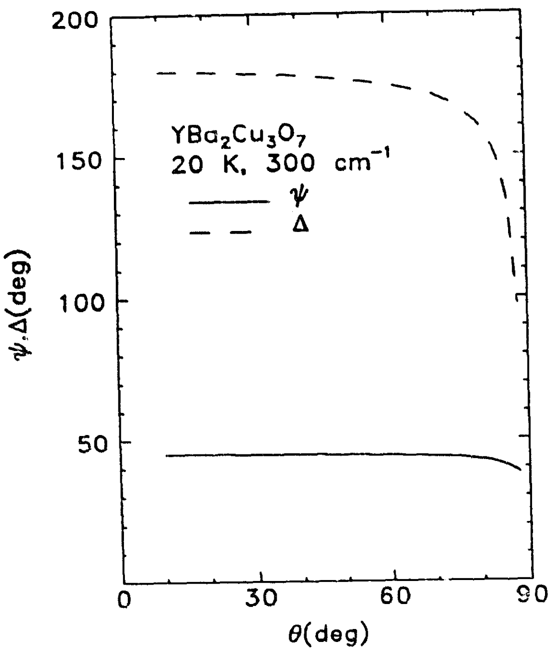

Figure 1. Ellipsometric angles for $YBa_2Cu_3O_{7-\delta}$ at 300 cm^{-1} vs. angle of incidence.

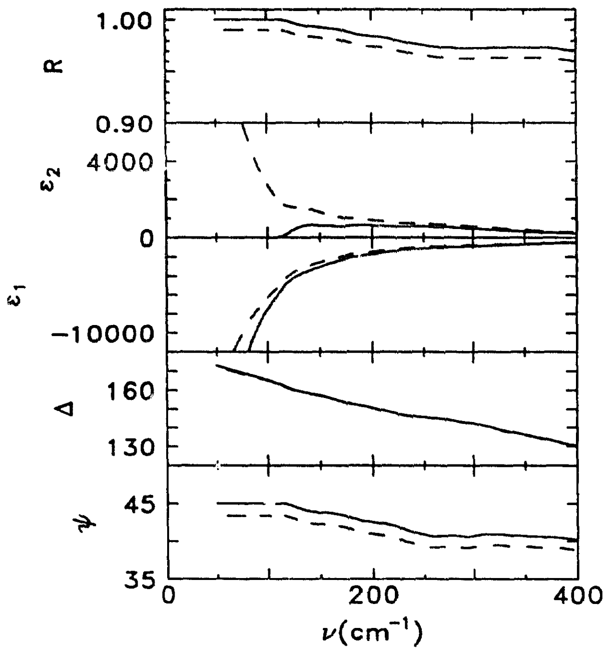

Figure 2. "Ideal" and "scaled" R, ε, Δ and ψ curves (see text).

cm^{-1}; the dashed lines were obtained after scaling this curve by 0.99. On both sets of reflectivity data, we performed a standard Kramers-Kronig analysis to obtain ϵ_1 and ϵ_2, respectively; then we proceeded with the calculation for an anisotropic medium as mentioned above, without adjusting the c-axis input, in order to get ψ and Δ. This way we obtain the variations in the ellipsometric angles causing the same error in the dielectric function as 1 percent in reflectivity: 0.4° in Δ and 1.6° in ψ in the region below 400 cm^{-1}, where a gap feature would be expected. These values are within or only slightly above the error bars corresponding to 1 degree variation in the angle of incidence, the lower limit of our convergence angle (0.7° for ψ and 7° for Δ, from the curves of Fig. 1). Thus the same problems are encountered in ellipsometry as in normal-incidence reflectivity when trying to define the fine details of ϵ_2 going to zero.

The effects of error propagation are far less dramatic on ϵ_1, whose low-frequency behaviour can be used for determining the penetration depth through the oscillator strength of the superfluid condensate [3]. Ellipsometry can give essential contributions in determining this quantity without the distorting effects of low-frequency extrapolations on Kramers-Kronig analysis.

This work has been supported by the European Community EE-EC grant No. ERB 3510 PL 922716.

REFERENCES

1. J. Humlíček *et al.*, Physica C 222 (1993) 166.
2. K.-L. Barth *et al.*, Thin Solid Films 234 (1993) 314.
3. K. Kamarás *et al.*, Phys. Rev. Lett. 64 (1990) 84.
4. C. C. Homes, unpublished.
5. R. M. A. Azzam and N. M. Bashara, *Ellipsometry and Polarized Light*, North-Holland, Amsterdam, 1977.
6. J. S. Kim *et al.*, Phys. Rev. B 49 (1994) 13065.