A cryostatic setup for the low-temperature measurement of thermal diffusivity with the photothermal method

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A cryostatic setup is described to perform photothermal deflection measurements from room temperature to 77 K. The setup uses gaseous nitrogen as a medium where the photodeflection is produced. The ability of the system to work is demonstrated presenting some measurements of thermal diffusivity of high-temperature superconductor samples and of yttrium-iron garnets with variable aluminum content. © *1995 American Institute of Physics.*

I. INTRODUCTION

The photothermal deflection (PD) technique is a well-known method for the measurement of thermal diffusivity of solid materials.[1] The contactless nature of the technique, its local nature, and the possibility of applying it to layered materials to obtain information on the film's thickness and on the presence of defects beneath the surface make it very useful in many cases.

By using specially designed furnaces, PD has also been used to study the thermal behavior of samples at a high temperature.[2]

Here we want to describe a special cryostatic measurement apparatus that has been implemented to perform measurements at a low temperature down to liquid-nitrogen temperature in order to allow the measurement of the thermal diffusivity of high critical temperature superconductors. The main objective in the design is to provide a suitable surrounding medium for the cooled samples in which the refractive index gradient, necessary for the deflection measurements, can be produced in a reliable and reproducible way.

In the following we first summarize the main aspects of PD; then we describe the system, and finally report on some preliminary measurements made on ferrites of the $Y_3Al_xFe_{5-x}O_{12}$ kind with a variable quantity of aluminum, and on the high superconductor YBCO.

II. BRIEF DESCRIPTION OF THE METHOD

The use of the photothermal deflection to measure thermal diffusivity has been amply described in the literature.[1–3] We give here only a brief summary of the relevant aspects involved in the system we will implement.

The physical principle involved in the technique is very simple. The sample is heated at the surface with a focalized laser beam (pump beam) that is amplitude modulated in time with a mechanical chopper. The absorbed radiation produces a temperature field T that propagates according to the Fourier equation

$$\nabla^2 T = \frac{1}{D}\frac{\partial T}{\partial t}, \tag{1}$$

where D is the thermal diffusivity related to the thermal conductivity k, the volume density ρ, and the specific heat c by the relation $D=k/\rho c$. In the so-called transverse configuration, the temperature distribution, generated in the surrounding medium above the surface of the sample, produces a refractive index distribution, which deflects a probe laser beam traveling parallel to the surface. In general, from the deflection of the probe beam the thermal properties of the sample can be derived. The angular deflection Φ of the probe beam is described by the equation

$$\Phi = \frac{1}{n}\frac{dn}{dT}\int_s \nabla_t T\, ds, \tag{2}$$

where n is the refractive index, dn/dT is the opthothermal coefficient of the medium in which the probe beam travels, and ∇_t is the gradient in the direction orthogonal to the propagation direction. Therefore the angular deflection can be decomposed into a tangential component Φ_t parallel to the sample surface, and a vertical component Φ_n orthogonal to sample surface.

Usually the tangential component is considered in a measurement because it has a better signal-to-noise ratio and is more independent of the surrounding medium parameters.

Roughly speaking, the photothermal deflection technique allows the measurement of the thermal diffusivity D from the time t_r taken by the thermal waves to travel the distance y between the axes of the pump and probe beams, through a relation of the kind

$$t_r \approx y^2/D. \tag{3}$$

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The phase $\varphi_t(y)$ of the tangential component of the photothermal deflection as a function of y is of the form

$$\varphi_t(y)=\frac{y}{l}+\varphi_0, \tag{4}$$

where φ_0 is a constant depending on the experimental conditions and l is a length with the meaning of an equivalent thermal diffusion length. In most cases,[3] l coincides with the thermal diffusion length given by

$$l_t=\sqrt{D/\pi f}, \tag{5}$$

where f is the chopper frequency. In the case of samples with thermal diffusivity lower than the diffusivity of the surrounding medium it is not so, and special care must be used to obtain the "true" diffusion length from Eq. (4). Relation (4) is valid, as it is in most cases, if the medium is homogeneous so that D does not change with y. From Eq. (4) the way to obtain the diffusivity is easily understood. It is sufficient simply to evaluate the phase of the transverse deflection by changing the relative distance y between pump and probe beams. If $l=l_t$ the proportionality between phase and offset y directly gives the thermal diffusivity from Eqs. (4) and (5). When the coefficient sample diffusivity is lower than the surrounding medium diffusivity a special procedure, as described in Ref. 3, must be used to obtain l_t from l.

Two possible geometries can be chosen for the probe beam. In one case the probe is made to travel parallel to the sample surface (skimming configuration) at some distance from it; this we will call the vertical offset z. When the sample has a specular surface, a different geometry can be used (called bouncing) in which the probe beam is made to beat the surface under some angle.

The skimming configuration poses some problems because the phase signal depends in this case also on z. However in some cases, as it is when the sample surface has some roughness, the skimming configuration is the only one which can be used. It can be shown however that the linear dependence of the phase from l is not affected by z, which only influences φ_0 in Eq. (4).

This brief summary of the technique is enough to understand how the method should be put into operation for low-temperature diffusivity measurements. A number of optical windows must be provided for the laser beams, and a suitable fluid should fill the cavity where the sample is contained, to provide the surrounding medium for the probe beam deflection. This fluid should be chosen to have a diffusivity lower than the sample one, and a good optothermal coefficient dn/dT.

III. CRYOSTAT DESIGN

A view of the used cryostat is shown in Fig. 1. It has a cylindrical shape and its main structure consists of two chambers thermally insulated from each other. A rod, located along the cylinder axis, allows one to put the sample inside the inner chamber, which is provided with four windows of BK7 glass to allow the passage of the pump and probe beams. By using thin glass plates (not shown in the figure) suitably realized in order to avoid optical interference effects, the two beams can be aligned inside the cryostat. A liquid-nitrogen reservoir surrounds the central sample tube and supplies cryogen via a capillary to a heat exchanger. During operation the "gravity-feed" flow of liquid nitrogen is controlled by the gas exhaust valve on the cryostat top plate. A window block is positioned directly beneath the heat exchanger and completely surrounds the sample position while allowing optical access to the sample through the window apertures. The heat exchanger is fitted with a 100 Ω platinum sensor and a heater for variable temperature operation in conjunction with a temperature controller. A nickel-copper thermocouple is also used in direct contact with the sample holder. In this way the temperature is stabilized within 0.5 K. The liquid nitrogen is thermally insulated from the outside through a shirt in which vacuum is made by the simultaneous action of a rotative and a turbomolecular pump.

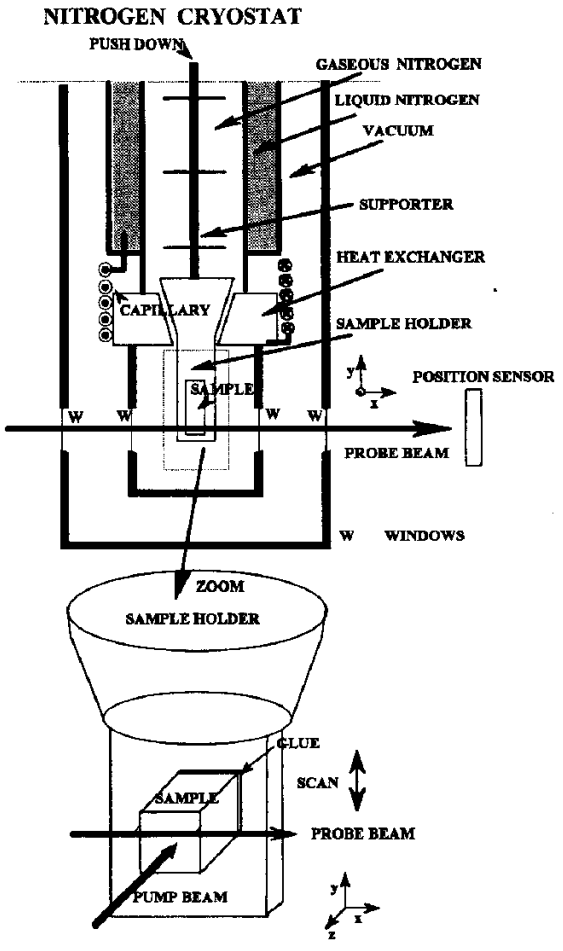

FIG. 1. Schematic of the cryostat.

The chamber containing the sample is filled with gaseous nitrogen which is the medium chosen to produce the deflection of the probe beam. Nitrogen has an optothermal coefficient dn/dT, which increases as the temperature lowers.[4]

The pump beam is obtained from a cw argon laser at 488 nm wavelength with a spot size of 0.75 mm. The output power can be varied from a few tens of milliwatts to 1 W, according to the nature of the sample, mostly depending on

its optical absorption and reflection coefficients at 488 nm. The probe beam is obtained from a cw He-Ne laser at 632 nm wavelength with a spot size of 0.4 mm. The power of this laser in most cases is fixed at 5 mW. Both pump and probe beams are expanded through a collimator made with two lenses with their focal plane in common. The pump beam is mechanically chopped at a frequency f that can be changed in the range from 1 to 1000 Hz. The possibility of changing the chopper frequency is very important because by varying this frequency the spatial scale on which the measurement is done can be varied [remember that the thermal diffusion length is a function of frequency (4)]. The temperature distribution which is produced in the sample, due to the heat diffusion process, can be seen as the result of the superposition of a field of thermal waves with an attenuation coefficient proportional to $\sqrt{f/D}$. It is therefore clear that for a given material, and therefore for a given value of D, by changing the frequency f, the extension of the heated zone can be varied. This is very useful, for example, when the sample is inhomogeneous and one wants to heat the sample on a length scale comparable with the dimension of inhomogeneities to obtain information on their dimension.

The gaseous nitrogen in the sample chamber is taken at a pressure between 200 and 1000 mbar in order to provide a sufficiently strong signal. Nitrogen has been chosen because it is very easy to obtain, it has no strong reactivity, it is easy to handle, and has a thermal diffusivity which decreases as temperature lowers. This property allows the use of Eqs. (3) and (4) without problems.

The deflection of the probe beam is measured by means of a position sensor in the form of a quadrant photodiode made by four photocells electrically connected in different ways according to whether a measure of the tangential or transverse deflection component is wanted. An interference filter is put in front of the photocells to select only the probe beam wavelength. The signal from the photodetector is sent to a lock-in amplifier which gives the modulus and phase of the deflection signal. The lock-in amplifier is synchronized at the chopper frequency.

To change the relative position of pump and probe beams one has chosen to maintain the fixed probe beam with its detection system and to translate instead the pump beam. Obviously this is a much simpler operation and it is accomplished through a mirror mounted on an electronic translational set connected with a control unit, which allows a spatial resolution of 1 μm.

The whole measurement system is interfaced with a PC which establishes the translational step and receives from the lock-in amplifier the results to memorize. Temperature variations of the order of a few millikelvin are enough to give a good signal-to-noise ratio, so that the procedure of heating the sample with the pump beam does not change appreciably the temperature of measurement which is known with a precision of 0.5 K, as already said.

IV. PRELIMINARY TESTS

The ability of the whole system to perform measurements at low temperature has been tested by using a molybdenum sample whose thermal properties are well known[5] and reproducible. The measurements have been performed at various temperatures and with various gaseous nitrogen pressures in the measurement chamber. In all cases thermal diffusivity values in agreement with the values reported in the literature have been found. To show the good working ability of the system, Figs. 2 and 3 show the behavior of the modulus and phase, respectively, of the transverse deflection signal as a function of the lateral offset y for various values of the gaseous nitrogen pressure between 200 and 750 mbar, at a fixed temperature of 78 K.

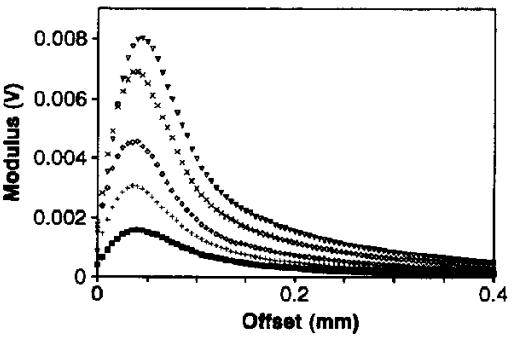

FIG. 2. Modulus of the tangential deflection on Mo sample as a function of pressure: (■) P=200 mbar, (+) P=340 mbar, (◇) P=600 mbar, (×) P=650 mbar, (▽) P=750 mbar. Chopper frequency f=625 Hz, T=78 K.

Simple inspection shows that by increasing the nitrogen pressure, the modulus of the signal also increases. This behavior is due to the dependence of the nitrogen refractive index n and dn/dT with pressure. This behavior is practically linear, as shown in Fig. 4, which gives the value of the modulus Φ_t for a given value of offset y, as a function of nitrogen pressure at a fixed temperature of 78 K.

The best signal-to-noise ratio is therefore obtained at low temperature and high nitrogen pressure. It must be remembered, however, that at a pressure P=1000 mbar (about 1 atm) and T=77.3 K, gaseous nitrogen tends to condense in droplets which can scatter the probe beam and avoid the measure of deflection. We have therefore chosen to perform our low-temperature measurements keeping the gaseous nitrogen pressure below 800 mbar.

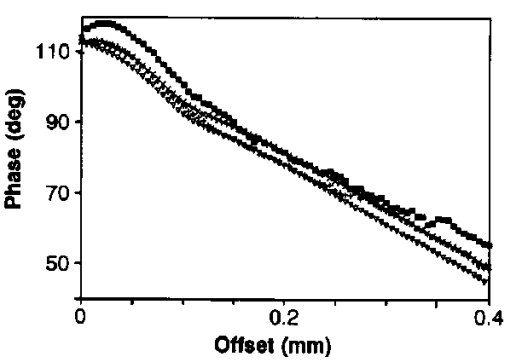

FIG. 3. Photothermal phase for a Mo sample at T=78 K as a function of offset y for different pressures: (■) P=200 mbar, (+) P=340 mbar, (×) P=650 mbar, (▽) P=750 mbar. Chopper frequency f=625 Hz. From these measurements a value of D=2.4 cm^2/s is derived.

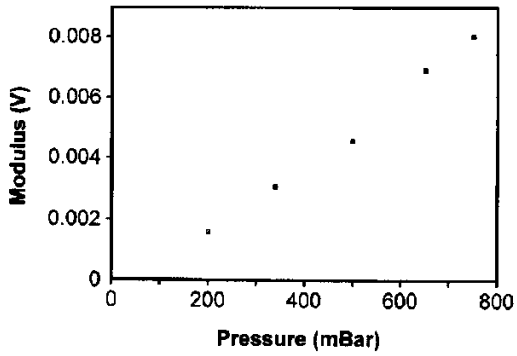

FIG. 4. Modulus of the tangential deflection as a function of nitrogen pressure for a Mo sample, for a given offset $y=0.045$ mm, $f=625$ Hz, $T=78$ K.

With respect to the phase of the signal, as shown in Fig. 3, by changing the nitrogen pressure the ratio $\Delta\varphi/\Delta y$ is the same. This assures one that the measurement of the thermal diffusion length is not affected by errors due to changes in the nitrogen pressure in the measurement chamber.

V. EXPERIMENTAL RESULTS

We report here preliminary measurements performed with the described setup. At first we will show the results obtained with some YBCO ($YBa_2Cu_3O_{7-x}$) samples. As is well known, YBCO is a material from the family of superconductor metallic oxides which has a high critical temperature around 92 K, and has assumed a great interest due to the possibility to obtain it in the form of a thin film. Due to its high transition temperature, our system allows one to study the behavior of its thermal diffusivity in the superconductor transition region.

Figure 5 shows the thermal diffusivity in the range between 80 and 105 K of a bulk YBCO sample. Starting from a high temperature, the diffusivity increases with decreasing temperature, shows some plateau around 90–96 K, followed by a sudden jump to higher diffusivity values, which after some leveling in the region between 83 and 89 K, increases again. Figure 6 shows some measurements made on another sample for which, near 92 K, there is a jump in diffusivity which, however, decreases at lower temperature to increase again by decreasing the temperature further.

A second example is given by measurements performed on magnetic garnets of the $Y_3Al_xFe_{5-x}O_{12}$ type. These ferrites have importance for microwave applications where they are used to implement isolators, gyrators, circulators, etc., which all are based on the nonreciprocal Faraday effect. In Fig. 7 the behavior of thermal diffusivity as a function of temperature is shown for garnets of four different compositions obtained by varying the aluminum concentration in the range $x=0-1.1$. Thermal diffusivity increases by a factor of ~ 20 at $T=78$ K with respect to its value at room temperature for the garnets free of aluminum and shows smaller increases as the aluminum content increases.

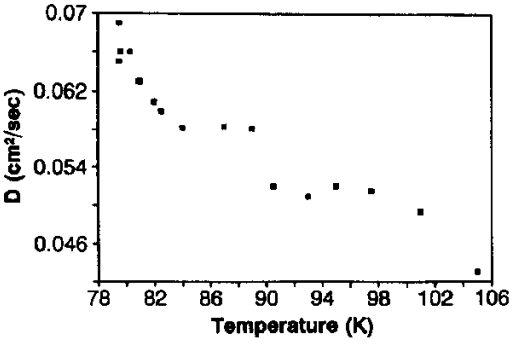

FIG. 5. Thermal diffusivity vs temperature for a YBCO sample.

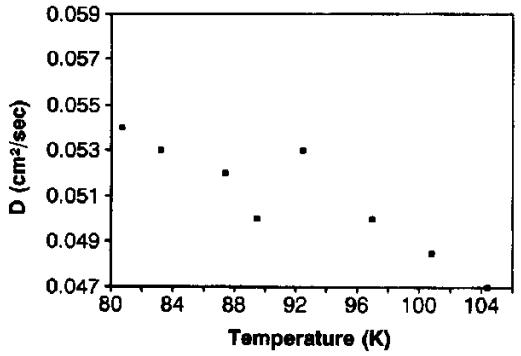

FIG. 6. Same as Fig. 5 for a different YBCO sample.

VI. DISCUSSION AND CONCLUSION

The experimental results shown above for YBCO and aluminum iron garnets show the good working of the assembled setup for the measurement of thermal diffusivity in the low-temperature range from room temperature down to 77 K. The results shown in Figs. 5 and 6 concerning YBCO can be interpreted considering that according to the standard CBS theory, the electron pairs produced below the critical temperature do not transport heat and do not scatter phonons.[6] The first property is responsible for the electronic contribution to heat transport tending exponentially to zero in the superconductive state. The second property produces instead an increase of the phonon contribution to heat transport, due to the increased mean free path of phonons. These two effects are directed in opposite directions and the material behavior depends on which mechanism is dominant. Generally a decrease of thermal conductivity is observed[6] due to the disappearance of the electronic contribution, as

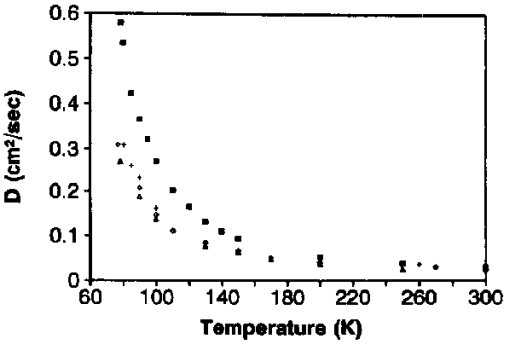

FIG. 7. Thermal diffusivity for garnets $Y_3Al_xFe_{5-x}O_{12}$ for different aluminum contents: (■) $x=0$, (+) $x=0.4$, (◇) $x=0.8$, (△) $x=1.1$.

TABLE I. Value of the mean free path for the garnets examined.

Molar fraction	$x=0$	$x=0.4$	$x=0.8$	$x=1.1$
$T=300$ K	12.5 Å	11.3 Å	10.8 Å	10.4 Å
$T=78$ K	241 Å	128 Å	129 Å	112 Å

can be observed in our Fig. 6. However if the heat transport is strongly limited by disorder, a thermal conductivity increase can be observed when the electronic contribution to heat conduction decreases, as seems to be the case in Fig. 5. The measurements of thermal diffusivity on the superconductor specimens allow, therefore, determination of, besides the value of diffusivity itself, the critical temperature range, and give information on the structural quality of the material.

The diffusivity increase observed for the garnets examined is due to the decrease of the phonon concentration and therefore of the phonon–phonon scattering rate which implies an increase of their mean free path. The length λ of the mean free path of phonons can be evaluated through the relation

$$K=\tfrac{1}{3}\,C_v\lambda V, \tag{6}$$

which connects the thermal conductivity K to the mean free path of phonons λ, the sound speed V, and the specific heat capacity per unit volume C_v. In particular, starting from the thermal diffusivity coefficient, the value of λ, for a given temperature, can be obtained by

$$\lambda=\frac{3K}{VC_v}=\frac{3D}{V}. \tag{7}$$

In Table I, the mean free path evaluated in all the samples examined for two different temperatures ($T=300$ K and $T=77$ K) is shown.

To obtain the values from Eq. (7), a value of $V=7.2\times10^5$ cm/s has been used.[8] The value obtained for $T=300$ is in good agreement with that found by Shchelkotunov.[7]

The phonon mean free path is limited by collisions with lattice defects and with other phonons so that

$$\frac{1}{\lambda}=\frac{1}{\lambda_{ph-ph}}+\frac{1}{\lambda_{\text{ph-def}}},$$

where $\lambda_{\text{ph-ph}}$ and $\lambda_{\text{ph-def}}$ refer, respectively, to phonon–phonon and phonon–defect scattering processes. The strong decrease of λ with the increase of x at low temperature ($T=78$ K) shows that $\lambda_{\text{ph-def}}$ becomes predominant, suggesting that the Al ions act as efficient scattering centers of phonons.

In this case again, besides the value of the thermal diffusivity, one has obtained some information on the structural behavior of the material.

Of course the same setup design can be implemented for measurements at lower temperatures, when necessary, by carefully choosing the surrounding gaseous medium (for example, helium).

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