

Superconducting Gap in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$

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(Received 25 May 1990)

Reflectivity measurements, at different fixed frequencies, have been performed on $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ films as a function of temperature and magnetic field. Analysis of the data provides evidence for an order parameter $2\Delta(0) \approx 3.3k_B T_c$ although the variation of this order parameter with temperature is significantly different from that predicted by the weak-coupling BCS model.

PACS numbers: 78.30.Er, 74.70.Vy, 78.20.Ci

The nature and exact value of the superconducting energy gap Δ in the high- T_c materials is still a matter of controversy. Despite the many techniques used, there is no general agreement on the observed ratio $2\Delta/k_B T_c$. Reported values^{1,2} range from 2 to 8. It has been argued³ recently that with the infrared reflectivity technique the gap would not be observable in the clean limit. This, as shown below, is indeed true and thus one should not select samples which are "too clean" for such experiments. Another fundamental issue is the variation of 2Δ with temperature close to T_c which has not been measured in any detail up to now.

The technique we used is to measure the reflectivity R at fixed frequencies as a function of the temperature T or the magnetic field B . It is found that $R(\omega_{\text{fixed}}, T \text{ or } B)$ is very sensitive to those excitations which are essentially dependent on the order parameter in a superconducting material and hence on T or B . This was already evidenced in previous measurements of the temperature-dependent transmission⁴ of V_3Si .

Photons of different frequencies are delivered by an ultrastabilized far-infrared laser specially developed⁵ for EPR measurements in high magnetic fields. The finite number of discrete lines spans an energy range from 1 to 37 meV. The incoming and reflected unpolarized beams are guided through light pipes and detection is made outside the cryostat. In this way the sensitivity of the detector is influenced neither by the (variable) temperature of the sample holder nor by the applied magnetic field. These measurements have been performed at normal incidence on superconducting films of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ (~ 300 nm thickness) deposited by dc sputtering⁶ on MgO substrates oriented along (100) and subsequently annealed. The c axis is perpendicular⁶ to the film plane and resistivity measurements give a superconducting transition temperature which depends on preparation and ranges from 78 to 87 K (zero resistance) with a width of the order of 7 K. A slightly different dc resistivity ρ_0 at 120 K is also obtained for different samples. When applied, the magnetic field was along the c axis.

Figure 1 summarizes the results we obtained on film A ($T_c \approx 87$ K, $\rho_0 \approx 3 \times 10^{-4}$ Ω cm) and film B ($T_c \approx 78$ K, $\rho_0 \approx 3 \times 10^{-4}$ Ω cm) for the order parameter 2Δ as a function of the temperature. Results at different magnetic fields B are also reported for sample A . Note that $2\Delta(0)$ at $B=0$ T is found to be of the order of $3.3k_B T_c$ for this film whereas the variation of 2Δ with T does not follow the weak-coupling BCS behavior near T_c .

These results at $B=0$ T have been deduced from the

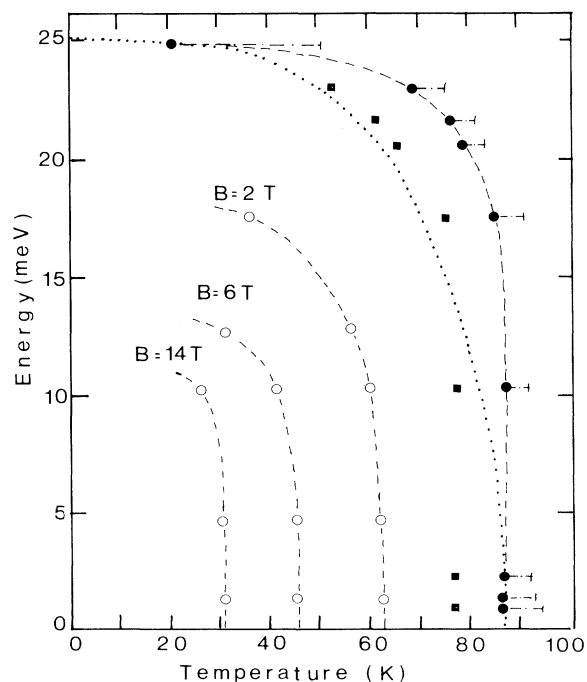


FIG. 1. Order parameter 2Δ as a function of temperature: Solid circles (sample A) and solid squares (sample B), without magnetic field; open circles (sample A), at different magnetic fields. The dashed lines are a guide for the eye. The dotted line is the weak-coupling BCS gap curve for $T_c = 87$ K. The dash-dotted lines give an estimate of the width ΔT (see text).

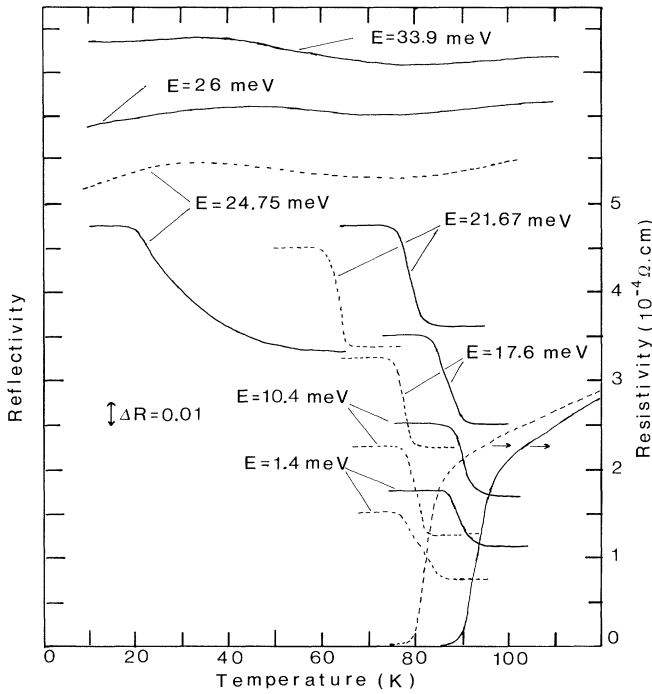


FIG. 2. Reflectivity traces (left scale) for different fixed energies as a function of temperature for sample *A* (solid lines) and sample *B* (dashed lines). The curves are displaced arbitrarily along the vertical axis for clarity. Traces of the corresponding resistivities (right scale) as a function of temperature for film *A* (solid line) and film *B* (dashed line).

various reflectivity traces (Fig. 2) obtained by scanning, at fixed frequencies, the temperature with a helium-flow cryostat. This scanning was performed at a rate of 1 K/min for increasing and decreasing temperatures without significant hysteresis. At low temperatures, R remains essentially constant. Then, depending on $\hbar\omega$, there is a critical temperature $T_1(\omega)$ above which R drops sharply and continuously until a second critical temperature $T_2(\omega)$ is reached. Above $T_2(\omega)$, R again remains smooth up to about 100 K. For frequencies equal to or higher than 26 meV for sample *A* and 24.75 meV for sample *B* no sharp drop in R is detected. Instead some smooth variation is observed. If we assume that for spectra obtained at lower frequencies the low-temperature reflectivity is close to 1, R can be calibrated

$$\sigma_{1s} = \frac{1}{2\omega} \int_{\Delta}^{\hbar\omega - \Delta} dE \{1 - 2f(E)\} G(E, E', q) - \frac{1}{\omega} \int_{\Delta + \hbar\omega}^{\infty} dE \{f(|E'|) - f(E)\} G(E, E', q), \quad (1)$$

where

$$G(E, E', q) = [g(E, E') - 1] \sigma_{1n}(|\epsilon| - |\epsilon'|, q) + [g(E, E') + 1] \sigma_{1n}(|\epsilon| + |\epsilon'|, q) \quad (2)$$

with σ_{1n} the normal-state conductivity, $g(E, E')$ the coherence factor, $f(E)$ the Fermi function, E and $E' = \hbar\omega - E$ the quasiparticle energies, and ϵ and ϵ' the normal-state energies ($\epsilon^2 = E^2 - \Delta^2$, $\epsilon'^2 = E'^2 - \Delta^2$). In Eq. (1), the first integral is nonzero only for $\hbar\omega > 2\Delta$ and the second integral, which vanishes at $T = 0$ K, gives the contribution of the thermally excited particles across the superconducting gap. The variation of σ_{1s} with ω depends significantly on the ex-

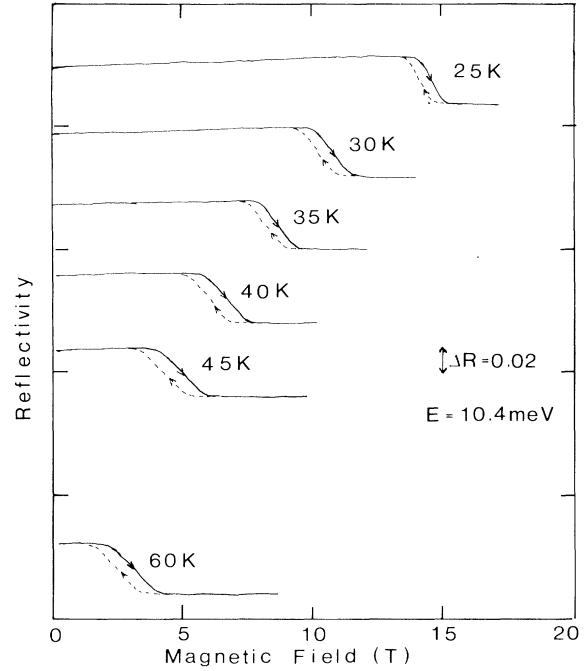


FIG. 3. Reflectivity traces at $E = 10.4$ meV and at different fixed temperatures for film *A* as a function of the magnetic field. Arrows indicate the direction of the magnetic-field sweep.

and the reflectivity drop ΔR quantified. The sharp onset at T_1 is found to always be reproducible, whereas ΔR varies within $\pm 0.5\%$ for different runs. The corresponding resistivity (right scale of Fig. 2) for each film shows an onset very close to T_1 (lower ω) except for very low frequencies where T_1 is 1–2 K lower than T_c .

As the magnetic field is swept at fixed T , the traces displayed in Fig. 3 were obtained for sample *A*. In that case, we observe some hysteresis, probably due to flux trapping. But ΔR is of the same order of magnitude as that observed when scanning the temperature. The data reported in Fig. 1 are those relating the different photon energies to the observed temperature T_1 or those extrapolated from magnetic-field measurements at a given B .

Within the framework of the BCS theory, Nam⁷ has provided a general formalism for the complex conductivity σ of superconductors. In the weak-coupling limit, the real part of σ reads

act form^{8,9} chosen for σ_{1n} . In the extreme anomalous limit σ_{1n} does not depend on ω , whereas in the $q \approx 0$ limit, $\sigma_{1n} = \sigma_0 / (1 + \omega^2 \tau^2)$ depends on the relaxation time τ in the simple Drude model. In all cases the reflectivity (calculated from σ_{1s} and its Kramers-Kronig counterpart σ_{2s} , with the appropriate sum rule⁸) has a sharp drop at $\hbar\omega = 2\Delta(T)$. In general, this drop is not expected to be so pronounced that one observes it experimentally in every case. However, when observable, it is related to the condition $\hbar\omega = 2\Delta(T)$. So, in Fig. 1, the curve at $B=0$ T represents indeed the variation of the order parameter measured with the electric field lying in the a - b plane of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ as a function of T . The curves, obtained for different magnetic fields, correspond to a pair-breaking effect⁷ which reduces $2\Delta(T)$. These magnetic-field effects will not be discussed further because their interesting aspects would require a more detailed experimental investigation. We, however, point out that the extrapolated values of $T_c(B)$, from $2\Delta(T_c, B) = 0$, fit exactly the onset temperature of finite resistance of film A at the corresponding magnetic field.¹⁰ So we are confident that the parameters displayed in Fig. 1 are related to the superconducting properties of the material.

It remains to explain the reflectivity drop $\Delta R(\omega) = R_s(\omega) - R_n(\omega)$ and the width $\Delta T(\omega) = T_2 - T_1$. Within BCS theory, both of these quantities are related to $\Delta(T)$ and the q and ω dependences of σ_{1n} in Eq. (1). In the extreme anomalous limit, R recovers its normal value R_n at $T = T_c$ which implies a ΔT increasing with ω . This is not observed, which is expected since these samples, having a coherence length much smaller than the mean free path, cannot be considered to be in the extreme anomalous limit. In the $q \approx 0$ limit, which seems more appropriate, σ_{1n} depends on two parameters σ_0 and τ . Results of the fit of the R traces versus T , at fixed ω , using Eq. (1) iteratively are presented in Fig. 4. Also shown in the inset is an example of a reflectivity trace obtained on a V_3Si crystal together with the theoretical curve assuming a BCS temperature variation of the gap¹¹ [$2\Delta(0) = 5$ meV, $\sigma_0 = 10000$ cm⁻¹, $1/\tau = 300$ cm⁻¹]. This agreement between experiment and theory for V_3Si gives convincing evidence that $\Delta(T)$ may be obtainable from $R(\omega_{\text{fixed}}, T)$. Note, however, that in the case of V_3Si $\Delta R(T)$ is rather broad.

It is clear from Fig. 1 that the fit of the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ $R(T)$ curves cannot be done using Eq. (1) when assuming the standard BCS temperature variation of the gap. We have then parametrized the dashed curve at $B=0$ T of Fig. 1 and used it in Eq. (1) [this parametrization gives $2\Delta(0) = 24.8$ meV for sample A]. Even so, the overall agreement is still poor in details. The discrepancy, at low frequencies, could perhaps be explained by an additional contribution of the fluctuations of Δ near T_c . However, the weak-coupling BCS model is clearly not appropriate at higher energies. The theoretic

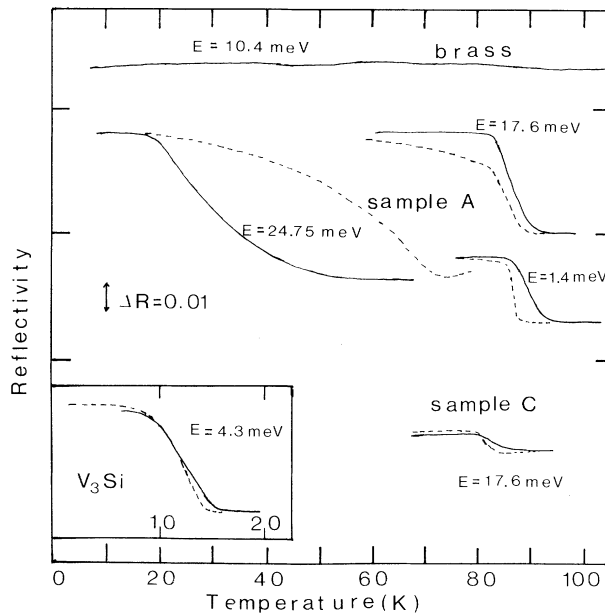


FIG. 4. Fit of the reflectivity traces of sample A and sample C . Solid lines are experimental results; dashed lines are theoretical fits using the model described in the text. Inset: Comparison of experimental and theoretical traces for V_3Si . Upper trace: Temperature run performed on a piece of polished brass in order to check the stability of the experimental assembly.

cal curves have been obtained with σ_0 varying between 10000 and 15000 cm⁻¹ and $1/\tau$ between 100 and 200 cm⁻¹ without being able to decide between equally poor fits. We have tried to use a temperature-dependent τ , as already proposed,¹² but without any significant improvement. Note that the discrepancy cannot be attributed to an experimental artifact since the trace obtained for a polished piece of brass (Fig. 4) does not exhibit major fluctuations over the same temperature range. So, the model itself appears inadequate. Our use of Eq. (1) involves many assumptions which could be questioned. These include the following, among others: (i) The overall formulation implies an isotropic superconductor treated in the framework of the mean-field theory; (ii) the k vector is assumed to be not conserved during the optical transition in the superconducting state; and (iii) in the normal state the simple Drude model is assumed. Some of them are clearly inadequate. We do not think that the inadequacy of the model alters the fact that we indeed measure 2Δ .

We have also performed experiments on a third sample C ($T_c \approx 83$ K, $\rho_0 \approx 8 \times 10^{-5}$ Ω cm) which essentially differs from the other two by a conductivity that is 3-4 times higher. As seen in Fig. 4, $\Delta R(17.6$ meV) for sample C is much smaller than the corresponding quantity for sample A , which agrees with theory and the arguments concerning clean samples already mentioned.³

This may be the reason why Reedyk *et al.*¹³ were not able to deduce a superconducting gap from their ω -scanned reflectivity data on bulk samples.

Finally, the smooth variation of $R(T)$ for energies slightly above $2\Delta(0)$ (Fig. 2) is also significant and reproduced theoretically. We did not observe any sharp drop near T_c which would have been indicative of the presence of a second higher-energy gap. We estimate the gap $2\Delta(0)$ to be of the order of 25 meV for sample A, that is, $2\Delta(0) = 3.3k_B T_c$. Comparing the gap obtained in the present experiment with other optical investigations, we find good agreement with results provided by the Raman technique,¹⁴ where a clear onset of gap excitations is seen beyond 200 cm^{-1} . However, these results disagree with values deduced from photoemission experiments.^{15,16} We also note that this value is in fairly good agreement with those deduced from earlier infrared reflectivity work on the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ compound¹⁷ or $\text{MBa}_2\text{Cu}_2\text{O}_{7-\delta}$ materials¹⁸ reporting gaps close to $3.5k_B T_c$. The fact that our value for $2\Delta(0)/k_B T_c$ is significantly lower than some of the previous ones reported in the literature raises an important unresolved issue. One possibility is the existence of more than one infrared- (IR-) active gap. Their detection could be dependent on the precise experimental configuration used. Another possibility, more unlikely in our opinion, is that the IR gap is a sensitive function of sample preparation. The present situation is further complicated by the observation that the feature associated with the higher-value gap appears to persist even at $T > T_c$ in several experiments.

In conclusion, we have been able, using the optical reflectivity technique at fixed frequencies, to derive the variation of the superconducting order parameter as a function of temperature with the electric field in the a - b plane of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$. It is found that the superconducting gap 2Δ is of the order of $3.3k_B T_c$ at low temperatures but deviates substantially from the BCS weak-coupling behavior near T_c . We emphasize that our magnetic-field measurements showed exactly the same magnitude reflectivity drop as in the temperature scan. Also a perfect agreement (to within experimental uncertainties) has been obtained between $T_c(B)$ measured by dc resistivity and that by reflectivity. These magnetic evidences together with our theoretical analysis strongly support the conclusion that we are indeed measuring the energy gap. It will be, of course, interesting to investigate by the same technique other materials like $\text{YBa}_2\text{Cu}_3\text{O}_7$ to see if the same behavior is observed, and if it is possible to quantify the anisotropy of the gap in these materials which can be prepared with the c axis lying either in the plane or perpendicular to the plane of the film.

Le Service National des Champs Intenses is a labora-

toire associé à l'Université Joseph Fourier de Grenoble. One of us (S.G.L.) acknowledges support by NSF Grant No. DMR 8319024, U.S. Department of Energy Contract No. DE-AC03-76SF00098, a Guggenheim Fellowship, and the University Joseph Fourier de Grenoble. We would also like to thank J. M. Louis for his help in the early stage of the experimental measurements. We are grateful to Dr. J. P. Senateur for providing us with the V_3Si samples.

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