Optical and Elastic Constants of Crystalline Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ by Brillouin Light Scattering Spectroscopy

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Room-temperature optical and elastic constants of crystalline Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ were determined using data extracted from Brillouin light scattering spectra. Refractive indices at a wavelength of 532 nm obtained from bulk peak linewidth versus frequency shift ratios range from 1.8 $\leq n \leq$ 2.0 for directions close to the crystallographic c-axis, while those determined from an expression relating the refractive index to elastic constants and Rayleigh surface wave velocity in the [010] direction on the (001) plane were found to be higher by $\sim$ 20%. Extinction coefficients at 532 nm for directions in proximity to the c-axis ranged from 0.03 $\leq$ $\kappa$ $\leq$ 0.1 and are consistent with values determined using optical interference but are several times smaller than those obtained in reflectance experiments. These discrepancies in index and extinction coefficient, also implicit to the optical penetration depth and dielectric function derived from the optical constants, are attributed to a reduction in surface mode frequencies and increased optical absorption due to the presence of surface roughness. A roughness-induced reduction in surface mode frequencies also appears to account for why the value of elastic constant $C_{44}$ = 20 $\pm$ 2 GPa obtained in the present work via measurement of bulk phonon modes is 25% larger than that previously reported.

INTRODUCTION

Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi-2212) is one of three commonly studied crystalline phases of the high-$T_c$ superconductor Bi$_2$Sr$_2$Ca$_{n-1}$Cu$_n$O$_{2n-1+\delta}$ (BSCCO). Similar to other members of the cuprate family, Bi-2212 possesses orthorhombic symmetry and has been described as micaceous due to its layered morphology, with a cleavage plane perpendicular to the crystallographic c-axis [1]. Bi-2212 is structurally complex, exhibiting birefringence [2] and incommensurability along the b and c crystallographic axes [3 4].

Many properties of Bi-2212, especially the electronic and structural properties, have been studied in detail [3 5], but the optical and elastic properties have received little attention. Measurements of the dielectric function at visible wavelengths are scant and, with the exception of one study [6], report only the real [7] or imaginary [8] part of this quantity in this region of the electromagnetic spectrum. Furthermore, there is significant variation in these values. Two independent measurements of refractive index at 532 nm have also been reported [9 10] but only in one case was the direction of light propagation specified [10]. A similar situation exists with regard to the elastic properties with only two groups reporting values of elastic constants. Room-temperature values for elastic constants were determined from Brillouin scattering measurements of surface mode frequencies with the assumption of hexagonal symmetry [11]. Ultrasonic studies on crystalline Bi-2212 report values for several elastic constants for the true orthorhombic symmetry, but were limited to temperatures 80 K $\leq T \leq$ 260 K [12 13]. It is therefore clear that additional studies of the optical and elastic properties of Bi-2212 are necessary.

In this paper, room-temperature refractive indices and extinction coefficients for three Bi-2212 crystals at the commonly-used wavelength of 532 nm are extracted from Brillouin spectra by measurement of acoustic mode peak frequency shifts and linewidths. This method for determining the optical constants of opaque materials is uncommon, particularly at a single wavelength. Knowledge of the refractive index and extinction coefficient is especially useful because they can be used along with Kramers-Kronig transformations to determine other optical properties. Elastic constant $C_{44}$ of crystalline Bi-2212 is also determined from Brillouin scattering measurements of bulk acoustic phonon modes.

EXPERIMENTAL DETAILS

Samples

Table I summarizes some physical properties and characteristics of the Bi-2212 crystals used in the present study. These (001)-oriented samples for Brillouin scattering experiments were obtained from parent crystals using mechanical exfoliation. This process was relatively straightforward for TC91 and TC90 as both exhibited the usual platelet geometry expected of Bi-2212 crystals with a cleavage plane perpendicular to [001]. In contrast, TC78 was somewhat irregularly-shaped which made exfoliation of an acceptable sample more difficult.
Table I: Critical temperature ($T_c$), approximate dimensions, and morphology of Bi$_2$Sr$_2$CaCu$_2$O$_{8+d}$ crystals used in the present work.

| Name  | $T_c$ [K] | Dimensions [mm$^3$] | Morphology |
|-------|----------|---------------------|------------|
| TC91  | 91       | 10 x 2 x 0.02       | Planar     |
| TC90  | 90       | 2 x 1 x 0.05        | Planar     |
| TC78  | 78       | 1 x 1 x 0.5         | Irregular  |

Brillouin Light Scattering

Brillouin scattering is the inelastic scattering of light by thermally excited acoustic phonons. For a backscattering geometry as used in the present work, conservation of energy and momentum for the scattering process yield the following expressions for surface and bulk phonon velocities, respectively [14]:

$$V_R = \frac{f_R \lambda_i}{2 \sin \theta_i}, \quad (1)$$

$$V_B = \frac{f_B \lambda_i}{2n}, \quad (2)$$

where $f_X$ ($X = R, B$, where $R$ and $B$ correspond to surface and bulk modes, respectively) is peak frequency shift, $\lambda_i$ is the incident light wavelength, and $n$ is the refractive index of the target material.

Brillouin scattering experiments were carried out in air at room temperature using a backscattering geometry with the set-up shown in ref. [15]. A single mode Nd:YVO$_4$ laser emitting at a wavelength of $\lambda_i = 532$ nm served as the incident light source. To minimize reflection losses, the polarization of the laser beam was rotated from vertical to horizontal by use of a half-wave plate. It was then passed through attenuating filters to reduce the power to $\sim$10 mW and subsequently focused onto the sample using a $f = 5$ cm lens with $f/# = 2.8$. Scattered light was collected and collimated by the same lens and focused by a $f = 40$ cm lens onto the entrance pinhole ($d = 300$ $\mu$m or $450$ $\mu$m) of a six-pass tandem Fabry-Perot interferometer which frequency-analyzed the scattered light. The free spectral range of the interferometer was set to values from 30 GHz to 150 GHz, with the lower ranges being used when higher resolution was required. It should be noted that the use of such a low incident light power level was necessary to avoid sample damage and thermal effects caused by optical absorption; trial runs at power levels of $\geq 20$ mW generated noticeable sample heating and/or damage. As a result, spectrum acquisition times of $\sim$ 20 hours were required and even in these circumstances only limited data could be obtained for TC90 and TC91 due to the low quality of the spectra collected from these two samples.

![Figure 1: Room-temperature anti-Stokes Brillouin spectra of TC78 collected at an incident angle of 30° and 40°](image)

RESULTS

Brillouin Spectra

Fig. 1 shows room-temperature spectra of sample TC78 collected at incident angles of 30° and 40°, where $B_1$, $B_2$, $B_3$ are bulk modes. Note: Peak due to Rayleigh surface mode not shown.

Table II shows the peak frequency shift ($f_B$), and full-width at half maximum ($\Gamma_B$) for $B_1$ and $B_3$ at various incident angles, obtained by fitting a Lorentzian function to each Brillouin peak. It is noted that 0.28 GHz, the measured $\Gamma$ value of the central elastic peak in the Brillouin spectrum, was subtracted from each fitted $\Gamma$ to remove the instrumental contribution to the overall linewidth. It was not possible to obtain analogous values associated with $B_3$ for TC90 and TC91 due to difficulty in fitting the peaks in the spectra of these samples, however $f_{B_3}$ and $\Gamma_{B_1}$ were obtained for an incident angle of 30° and 65°, respectively. Peak $B_2$ was omitted from the analysis because its asymmetric shape (see Fig. 1) and abnormally large width ($> 2.50$ GHz) suggest that it may result from the superposition of two or more closely-spaced peaks.
Table II: Bulk acoustic mode frequency shifts ($f_{B_i}$) and associated linewidths ($\Gamma_{B_i}$) obtained from Brillouin spectra of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ samples used in the present study. $T_c$ - critical temperature; $\theta_i$ - incident angle.

| Sample | $T_c$ [K] | $\theta_i$ [deg] | $f_{B_1}$ [GHz] | $\Gamma_{B_1}$ [GHz] | $\Gamma_{B_1}/f_{B_1}$ | $f_{B_2}$ [GHz] | $\Gamma_{B_2}$ [GHz] | $\Gamma_{B_2}/f_{B_2}$ |
|--------|-----------|------------------|-----------------|----------------------|-----------------------|-----------------|------------------|------------------------|
| TC91   | 91        | 65               | 14.6±0.5        | 1.3±0.5              | 0.09                  | -               | -                | -                      |
| TC90   | 90        | 30               | 14.6±0.4        | 1.2±0.5              | 0.08                  | -               | -                | -                      |
| TC78   | 78        | 10               | 13.2±0.3        | 1.2±0.3              | 0.09                  | 31.9±0.3        | 1.1±0.3          | 0.03                   |
|        |           | 30               | 13.5±0.3        | -                    | -                     | -               | -                | -                      |
|        |           | 40               | 13.0±0.3        | 1.7±0.3              | 0.1                   | 32.6±0.3        | 1.0±0.3          | 0.03                   |
|        |           | 50               | 14.1±0.3        | 1.8±0.3              | 0.1                   | -               | -                | -                      |
|        |           | 60               | 13.4±0.3        | -                    | -                     | 33.3±0.3        | 1.1±0.3          | 0.03                   |
|        |           | 70               | 14.0±0.3        | 1.6±0.3              | 0.1                   | -               | -                | -                      |

Figure 2: Rayleigh surface mode peak frequency shift versus sine of incident angle for samples TC91 (□), TC90 (▲) and TC78 (○). Horizontal error bars are approximately the symbol width.

**Determination of Refractive Index and Extinction Coefficient**

*Linewidth-Peak Frequency Shift Ratio*

The refractive index ($n$) and extinction coefficient ($\kappa$) are related to the bulk Brillouin peak linewidth, $\Gamma_B$, and frequency shift, $f_B$, through the equation

$$\frac{\Gamma_B}{f_B} = \frac{2\kappa}{n}. \quad (3)$$

To obtain the refractive index and extinction coefficient of crystalline Bi-2212, a function was created containing each set of peak frequency shift and linewidth data for both Stokes (S) and Anti-Stokes (AS) Brillouin peaks:

$$\sum_{j=1}^{N} \sum_{i=1}^{3} w_{ij}^{(S)} \left[ \frac{\Gamma_{ij}^{(S)}}{f_{ij}^{(S)}} - \xi \right]^2 + w_{ij}^{(AS)} \left[ \frac{\Gamma_{ij}^{(AS)}}{f_{ij}^{(AS)}} - \xi \right]^2 = 0, \quad (4)$$

where $i$ and $j$ denote the $i^{th}$ bulk mode of the $j^{th}$ data set, $\xi = 2\kappa/n$ and $w_{ij}$ is the function weight of each component. Due to the uncertainty in frequency shifts and widths being the same for a given sample (see Table II), the corresponding $w_{ij}$ were constant.

By substituting sets $f_i$ and $\Gamma_i$ from Table II for each sample into Eq. (4) along with the initial guesses for $n$ and $\kappa$ ranging from 1.4−3.0 and 0.01−1.00, respectively (chosen based on previous estimates of these constants in the visible region of the electromagnetic spectrum [6, 9, 10]), Eq. (4) was minimized using the sequential least squares programming optimization method (SLSQP). Given that Eq. (4) is a series of ratios which may possess an infinite set of solutions, $n$ and $\kappa$ were constrained to 1.0−6.0 and 0.01−1.00, respectively, with the set that minimizes the function defined in Eq. (4) taken as the final values. This process yielded 1.9 and 0.09 for TC91, 1.8 and 0.08 for TC90, and 1.9 and 0.07 (averaged) for TC78 for $n$ and $\kappa$, respectively. Eq. (4) was plotted as functions of $n$ and $\kappa$ to ensure the function itself is not flat and has a pronounced minimum at the quoted values.

**Equation for Rayleigh Surface Wave Velocity**

A second estimate of the refractive index can be extracted from the Brillouin data via an implicit equation for the Rayleigh surface acoustic wave velocity, $V_R$, in the [010] direction on the (001) plane for a crystal with orthorhombic symmetry [17].
\[
\sqrt{1 - \frac{V_T^2}{V_i^2}} \left[ 1 - \frac{C_{23}^2}{C_{22}C_{33}} - \frac{\rho V_T^2}{C_{22}} \right] = \frac{\rho V_T^2}{C_{22}} \sqrt{C_{33} \left[ 1 - \frac{\rho V_T^2}{C_{22}} \right]}, \tag{5}
\]

where the \(C_{ij}\) are elastic constants, \(\rho = 6510\, \text{kg/m}^3\) is the density, and \(V_T\) is the velocity of the slow quasi-transverse mode propagating in the [001] direction. In a Brillouin scattering experiment, this velocity is given by \(V_T = f_B \lambda_i / 2n\) as dictated by Eq. 2, which upon substitution into Eq. 5 yields

\[
n = \frac{f_B \lambda_i}{2V_R} \sqrt{1 - \frac{(\frac{C_{22}}{C_{33}}) [\alpha - 1] - \beta}{\alpha - \beta}}^2, \tag{6}
\]

where \(\alpha = 1 - \rho V_T^2 / C_{22}\) and \(\beta = C_{23}^2 / C_{22}C_{33}\).

As shown in Fig. 2, the Brillouin data give velocities \(V_R = 1410\, \text{m/s}, 1570\, \text{m/s},\) and \(1610\, \text{m/s}\) for TC78, TC91, and TC90, respectively. These velocities are for unknown directions of propagation on the [001] plane, but it has been shown that the Rayleigh surface mode velocity on this plane is essentially constant \([11]\). The measured velocities are therefore taken as equal to those along the [001] direction for the purposes of computing \(n\).

Elastic constants \(C_{22}\) and \(C_{33}\) have been determined for crystalline Bi-2212 with \(C_{22} = 110\, \text{GPa}\) at 260 K, the nearest temperature to ambient for which a value of this constant has been reported \([13]\), and \(C_{33} = 75.8\, \text{GPa}\) at room temperature with the approximation of hexagonal symmetry \([11]\). No value of \(C_{23}\) appears in the literature, but because the symmetry of Bi-2212 can be approximated as tetragonal (since \(a \approx b\) \([3]\), \(C_{23} \approx C_{13}\), with \(C_{13} = 56\, \text{GPa}\) \([11]\).

Using the \(V_R\) and \(C_{ij}\) values given above and frequency shifts \(f_B\), from Table \([1]\), the refractive indices of the three samples of the present work as determined from Eq. 6 are \(n = 2.4\) for TC78, \(n = 2.3\) for TC91, and \(n = 2.2\) for TC90. It should be noted that the frequency shifts \(f_B\) were obtained from spectra collected at nonzero angles of incidence and therefore the probed phonons were not travelling along the [001] direction. For angles ranging from 5° to 30° from the [001] direction, however, the shifts were found to be largely independent of direction and therefore the shift value obtained from the spectrum collected at the lowest angle of incidence was taken as being equal to that along the [001] direction. These angles are indicated in Table \([3]\).

## DISCUSSION

### Optical Properties

Table \([1]\) shows the optical constants, \(n\) and \(\kappa\), at 532 nm obtained using the methods described above, along with derived quantities and previously published values. The refractive index values obtained using the linewidth-shift ratio range from 1.8 to 2.0 and are within ≤ 10% of those determined using a combined reflectance-ellipsometry method \([6]\), reflectance spectroscopy \([10]\), and optical interference \([9]\). In contrast, Eq. \([6]\) yields values that are ~ 20% larger than previously published results, with 2.2 ≤ \(n\) ≤ 2.4. This large difference could be due to a reduction in surface mode frequencies resulting from surface damage and/or roughness \([19, 20]\). This results in a decrease in surface phonon velocity \(V_R\) and consequently an increase in \(n\) determined by Eq. \([6]\).

More specifically, if surface roughness is present, \(n\) obtained from Eq. \([6]\) should be larger than that determined from a measurement that probes bulk phonons within the volume of a material, like the linewidth-shift ratio. This is precisely what is observed. As an example, consider the case of TC78, the sample with the roughest surface as confirmed by optical microscopy and that for which \(V_R\) gives the highest \(n\) value. If \(V_R = 1410\, \text{m/s}\) or TC91 (1570 m/s) is used instead of \(V_R = 1410\, \text{m/s}\) for this sample in Eq. \([6]\) (given the pristine (001) surface of TC90 and TC91 due to a high-quality cleave), one obtains \(n = 2.0 – 2.1\). These values are within ~ 10% of those determined using the linewidth-shift ratio.

Extinction coefficients obtained using the linewidth-shift ratio range from 0.03 ≤ \(\kappa\) ≤ 0.1. Values for \(\kappa\) corresponding to the refractive indices determined from Eq. \([6]\) were also obtained via the numerical values of ratio \(\Gamma / f\) given in Table \([1]\). The extinction coefficients obtained in the present work are similar to that found by optical interference but are, at minimum, ~ 50% smaller than those obtained via reflectance analysis \([6, 10]\). Such high \(\kappa\) values result in 2\(\kappa / n\) ratios that would require, via Eq. \([5]\), linewidth-shift ratios \(\Gamma_{B_i} / f_{B_i}\) several times larger than those obtained in the present Brillouin scattering experiments (see Table \([1]\) and also those extracted from spectra shown in previous independent Brillouin studies of Bi-2212 \([11]\). As for the elevated \(n\) values from Eq. \([6]\), surface roughness could account for this discrepancy in \(\kappa\) values obtained by Brillouin scattering and reflectance as it is known to result in increased optical absorption \([21, 22]\). Such roughness would be manifested in the reflectance result because it is primarily a measurement of light reflected from the sample surface. In contrast, surface roughness will not likely impact the bulk Brillouin result because the probed bulk phonons propagate in the volume of the material.

Table \([11]\) also shows values for the optical penetration
depth and dielectric function at 532 nm derived from the optical constants. The penetration depth, $d = \lambda_i/4\pi\kappa$, determined from optical constants obtained using the linewidth-shift ratio and Eq. 5 are, in general, quite similar, averaging 460 nm and 440 nm, respectively. These values are $\sim 2-4$ times larger than those previously reported [6,10] due to the above-noted differences in $\kappa$. The dielectric function ($\tilde{\epsilon} = \epsilon_1 + i\epsilon_2$) at 532 nm was determined using the relationship between dielectric and optical constants: $\epsilon_1 = n^2 - \kappa^2$ and $\epsilon_2 = 2n\kappa$. On average, $\epsilon_1$ values determined using optical constants found from the linewidth-shift ratio are similar to those obtained in other studies [6,9,10] but are $\sim 35\%$ smaller than those determined using constants found from Eq. 6, primarily due to the higher value of $n$ supplied by this equation. Values for $\epsilon_2$ calculated from optical constants determined using the linewidth-shift ratio are also $\sim 35\%$ smaller on average than those determined using constants based on Eq. 6, but are several times smaller than $\epsilon_2$ values obtained in previous reflectance studies [6,10], mainly due to the higher $\kappa$ values obtained in the latter studies. These observations on the relative magnitudes of the optical penetration depth and dielectric function as determined by various means implicitly incorporate the trends in $n$ and $\kappa$ noted in previous paragraphs and therefore serve to highlight the effects of surface roughness on these derived quantities.

### Elastic Constant $C_{44}$

An important consequence of the presence of surface roughness is that measurement of a quantity in the surface region will yield a different value than in the bulk. In view of this, elastic constant $C_{44}$ for Bi-2212 was determined using quantities derived solely from measurements of bulk phonons to mitigate the impact of surface effects. For this calculation, results for the refractive index ($n = 2.0$) determined from the linewidth-shift ratio and the frequency shift ($f_T = 13.2$ GHz) for the peak due to the slow quasi-transverse bulk mode propagating nearly along the $c$-axis for TC78 were used due to the superior quality of the Brillouin data obtained from this sample relative to TC90 and TC91. Substitution of these values into the expression $C_{44} = \rho V_T^2 = \rho(f_T\lambda_i/2n)^2$ gives $C_{44} = 20 \pm 2$ GPa. This result is in accord with $C_{44} = \rho V_T^2 = 21.8$ GPa estimated using $V_T = 1830$ m/s from neutron scattering measurements [24,25], but $\sim 25\%$ larger than the lone published value of 15.8 GPa [11]. An analogous calculation for TC90 and TC91 yielded estimates for $C_{44}$ that, while crude due to the poor quality spectral data for these samples and because the direction of light propagation is at an appreciable angle to the $c$-axis, are also considerably larger than the previously published value.

In order to identify the mechanism responsible for the difference between the value of $C_{44}$ obtained in the present work and that previously reported, it is noted...
that the latter was determined from Brillouin scattering measurements of surface mode frequencies \[11\]. As noted above, surface roughness or damage can result in a reduction in frequencies of surface modes measured by Brillouin scattering \[18–20\]. Furthermore, it has also been stated that these frequencies might be low because elastic constants at the surface differ from those in the bulk \[14\]. The results obtained here are consistent with this premise and, in fact, the presence of surface roughness on crystalline Bi-2212 has been noted in another independent Brillouin scattering study \[24\]. Moreover, using \( n = 2.4 \) as found from Eq. 6, which contains surface mode velocity \( V_R \) and elastic constants \( C_{13} \) and \( C_{33} \) determined using surface mode frequencies \[11\], gives \( C_{44} = 14 \) GPa, a value within \( \sim 10\% \) of that quoted in ref. \[11\]. It therefore seems likely that surface roughness is responsible for the low value of \( C_{44} \) reported in an earlier study relative to that obtained in the present work.

**CONCLUSION**

Room temperature optical constants of crystalline Bi-2212 were determined at a wavelength of 532 nm using two approaches based on data obtained from Brillouin light scattering spectra. Optical constants determined from measurements of bulk phonons propagating within the volume of the crystal are, in general, noticeably different from those extracted from measurements of surface modes. These differences are attributed to a surface roughness-induced lowering of surface acoustic mode frequencies, which also appears to account for why elastic constant \( C_{44} \) determined in the present study is \( \sim 25\% \) higher than that previously reported.

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