Elastic constants of a monocrystal of superconducting $YBa_2Cu_3O_{7-\delta}$

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All nine independent elastic constants c_{ij} of a $YBa_2Cu_3O_{7-\delta}$ monocrystal were determined at room temperature using resonant ultrasound spectroscopy. Shear and Young moduli obtained from the c_{ij} by the Voigt-Reuss-Hill averaging method agree well with the pulse-echo results on a polycrystal. The Debye temperature calculated from the elastic constants agrees with the specific-heat measurement.

I. INTRODUCTION

A material's elastic properties are important because they relate to such various fundamental solid-state phenomena as specific heat, Debye temperature, and Grüneisen parameter. For superconductors, elastic constants can be linked explicitly to the superconducting transition temperature T_c through the Debye temperature Θ_D and the electron-phonon coupling parameter λ.[1]

Since the discovery of the high-T_c superconductor $YBa_2Cu_3O_{7-\delta}$ (YBCO) in 1987, its elastic constants have been studied extensively. Most of the studies, however, were carried out on polycrystalline specimens because existing monocrystals are too small for the usual measurement methods. Not surprisingly, the studies on polycrystals have many problems. For example, the reported bulk modulus varies from 27 to 196 GPa.[1] Compared with the number of studies on polycrystals, only a few elastic studies of monocrystals of orthorhombic YBCO were reported.[2–9] Among these studies, only Reichardt *et al.*,[6] using inelastic neutron scattering, reported a complete set of measured c_{ij} . Because they assumed tetragonal symmetry, they obtained only six of the nine independent c_{ij}.

Recent enhancements of resonant ultrasound spectroscopy[10,11] (RUS) enabled us to measure a small monocrystal specimen of YBCO and determine the elastic constants from the resonant frequencies. In this paper, we report the nine c_{ij} of orthorhombic YBCO measured by resonant ultrasound spectroscopy.

II. MATERIAL AND MEASUREMENTS

The specimen used for this study was a detwinned monocrystal of $YBa_2Cu_3O_{7-\delta}$, with $\delta < 0.1$. The crystal was grown in a gold crucible using a self-flux method. On crystals grown in the same way it was shown that about 10% of the chain copper atoms were replaced by gold. A comparison[12] of the reversible magnetization of crystals grown in gold crucibles and zirconia crucibles indicated that the superconducting properties near T_c were not affected by the gold impurities, whereas normal-state transport properties were known to change. The magnetic-susceptibility curve indicated that the superconducting transition occurred at 91.4 K with a transition width of 0.7 K. The specimen was cut into a rectangular parallelepiped with the edges aligned along the principal axes of the orthorhombic cell. The dimensions were $a = 1.435$ mm, $b = 1.164$ mm, and $c = 0.265$ mm. From the mass and the dimensions, the mass density was determined to be $\rho = 6.333$ g/cm^3, consistent with the theoretical value of 6.393 g/cm^3.

The resonant frequencies were measured using a resonant-ultrasound technique described in detail elsewhere.[10,11] Two $LiNbO_3$–diamond-composite piezoelectric transducers contacted the parallelepiped-specimen corners as shown in the inset of Fig. 1. The accuracy of the frequency measurements for this speci-

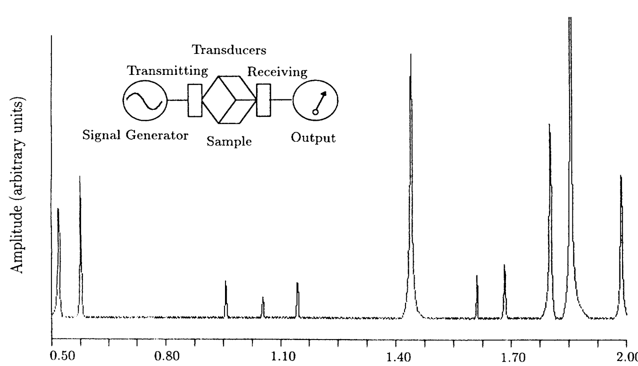

FIG. 1. Resonant-frequency spectrum for the specimen of $YBa_2Cu_3O_{7-\delta}$ monocrystal at 295 K. Inset shows the schematic diagram of our measurement setup.

men was about 30 ppm. Figure 1 shows the resonant-frequency spectrum for the specimen at room temperature from 0.5 to 2 MHz. For the analysis here, we used the data from 0.5 MHz to 3.5 MHz where a total of 30 resonances were observed.

III. RESULTS AND DISCUSSION

The resonant frequencies of a solid can be determined from its elastic constants c_{ij} using a method developed by Demarest[13] and improved by Ohno[14] and Visscher,[15] a straightforward procedure. However, it is very difficult to deduce elastic constants from resonant frequencies because no analytical method exists.

We solved the inverse problem in the following way. First, we estimated the elastic constants as closely as possible from other sources (for the present study, we started with the c_{ij} estimated by Ledbetter and Lei[17]). Then we calculated the resonant frequencies using the estimated elastic constants and the dimensions and mass density of the specimen.

Second, we defined a figure of merit,

$$F = \sum_{i=1}^{N} w_i \left(f_i - g_i\right)^2 . \qquad (1)$$

Here, g_i and f_i indicate the ith measured and calculated frequencies, respectively; N denotes the total number of measured frequencies; and w_i is a weighting factor chosen (usually either 0 or $1/g_i^2$, so that F is a measure of fractional deviation) to reflect one's degree of confidence in the measured frequency g_i.

Finally, we used the Levenberg-Marquardt method[18] as a systematic scheme to locate the minimum of F in a multidimensional elastic-constant space (a nine-dimensional space in this case). We estimated the accuracy of the elastic constants c_{ij} by considering the shape of the surface F near the minimum. F is assumed to be a quadratic function of the elastic constants in this neighborhood, and so the surfaces of constant F are ellipsoids with major axes related to the accuracy with which the corresponding elastic constants are determined. We estimated the accuracy for each c_{ij} by finding the length of the corresponding semimajor axis of the ellipsoid when F exceeds the minimum by 2%.

Table I shows elastic constants for monocrystals of YBCO obtained by various measurement methods and estimation. The last row contains our results. The root-mean-square error between the measured and calculated resonant-frequency spectra is less than 0.3%. The largest error in c_{ij} is estimated to be 7% for c_{33}. For three longitudinal moduli c_{11}, c_{22}, and c_{33} and the in-plane shear modulus c_{66}, there is a reasonable agreement among results reported by different groups (except for the c_{66} of Zouboulis *et al.*[9]). On average, our c_{ij} differ from those of Reichardt *et al.* by 17% and from those of Ledbetter and Lei[17] by 15% (omitting c_{12}). Compared with the values obtained by other groups, our c_{12} is high. Because no wave speed in the crystal depends only on c_{12}, it is no way to estimate it directly.

Besides comparisons with other results on monocrystals, we can also compare our c_{ij} with polycrystal measurements. Using a Voigt-Reuss-Hill method,[19] we obtained the quasipolycrystal elastic constants from our c_{ij}. They are B = bulk modulus = 138.2 GPa, E = Young modulus = 154.3 GPa, G = shear modulus = 58.70 GPa, and ν = Poisson ratio = 0.314. The Young and shear moduli agree well with the pulse-echo measurements on a polycrystal corrected to a void-free state reported by Lei and Ledbetter:[1] $E = 148.1$ GPa and $G = 58.99$ GPa. But the bulk modulus is higher: 138.2 versus 100.9 GPa. Using an ionic-crystal-model calculation and other measured physical properties, Ledbetter and Lei[20] concluded that the bulk modulus for YBCO equals 107 ± 10 GPa. Compared with the best bulk modulus, 115 GPa, measured on a monocrystal of YBCO using high-pressure x-ray diffraction by Aleksandrov, Goncharov, and Stishov,[2] our bulk modulus is higher by 20%. The reason for this might be associated with our determination of c_{12}.

We can also examine our c_{ij} by calculating the Debye temperature from the c_{ij} and comparing it to specific-heat measurements. The Debye temperature Θ_D can be

TABLE I. Monocrystal elastic constants for YBCO superconductor. Units are in GPa. B represents bulk modulus.

Source	c_{11}	c_{22}	c_{33}	c_{44}	c_{55}	c_{66}	c_{12}	c_{13}	c_{23}	B	Remarks
Aleksandrov, Goncharov, and Stishov[a]										115	X-ray diffraction
Baumgart *et al.*[b]	211		159	35							Brillouin spectroscopy
Golding *et al.*[c]	234[d]		145[d]								Ultrasound
Reichardt *et al.*[e]	230	230	150	50	50	85	100	100	100	131	Neutron scattering
Saint-Paul *et al.*[f]			160	25		82	66				Ultrasound
Zouboulis *et al.*[g]				42	33	57					Brillouin scattering
Jorgensen *et al.*[h]										123[i]	Neutron diffraction
Ledbetter and Lei[j]	223	244	138	61	47	97	37	89	93	115	Estimate
Present work	231	268	186	49	37	95	132[k]	71	95	138	Resonant ultrasound

[a]Reference 2.
[b]References 3 and 4.
[c]Reference 5.
[d]Measured at 80 K.
[e]Reference 6.
[f]References 7 and 8.
[g]Reference 9.
[h]Reference 16.
[i]Measured on a powder specimen.
[j]Reference 17.
[k]For this value see discussion in the text.

determined from the relationship:[21]

$$\Theta_D = \frac{h}{k}\left(\frac{3N_0}{4\pi V}\right)^{1/3} v_m. \tag{2}$$

Here, h and k denote the Planck and Boltzmann constants; N_0 indicates the number of independent three-dimensional oscillators in a volume V; and v_m denotes mean sound velocity defined by

$$v_m^{-3} = \frac{1}{M}\sum_{i=1}^{M}\frac{1}{3}\left(v_{\ell i}^{-3} + 2v_{ti}^{-3}\right). \tag{3}$$

Here, $v_{\ell i}$ and v_{ti} indicate longitudinal and transverse velocities in the ith propagating direction, respectively; M is the number of all propagating directions. For the present study, we chose $M = 70$. v_ℓ and v_t can be determined from the c_{ij} and density by solving the usual Christoffel equations[21] in a certain propagating direction. The Debye temperature determined from our c_{ij} is $\Theta_D = 414$ K, in a good agreement with the specific-heat value $\Theta_D = 440$ K.[22]

To conclude, we reiterate that for orthorhombic $YBa_2Cu_3O_{7-\delta}$ we determined a complete set of monocrystal elastic constants c_{ij} from the measured resonant frequencies. This set, except for c_{12}, agrees with the inelastic-neutron-scattering results and the estimation based on mainly monocrystal measurements. It also agrees well with polycrystal measurements of the Young modulus, shear modulus, and Debye temperature.

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