## JOURNAL OF APPLIED PHYSICS

## VOLUME 38, NUMBER 6

MAY 1967

## The Voigt-Reuss-Hill Approximation and Elastic Moduli of Polycrystalline MgO, CaF<sub>2</sub>, g-ZnS, ZnSe, and CdTe

D. H. CHUNG AND W. R. BUESSEM

#### Materials Research Laboratory, The Pennsylvania State University University Park, Pennsylvania

(Received 15 December 1966; in final form 23 January 1967)

The Voigt-Reuss-Hill (VRH) approximation, a useful scheme by which anisotropic single-crystal elastic constants can be converted into isotropic polycrystalline elastic moduli, is shown to apply for moderately anisotropic cubic crystals like MgO,  $CaF_2$ ,  $\beta$ -ZnS, ZnSe, and CdTe. Experimental values of polycrystalline isotropic elastic moduli for these materials are presented here, and the validity of the VRH approximation is established. The VRH approximation is then discussed for these materials with respect to their elastic anisotropy of crystals. To provide further support to this work, a numerical confirmation on the VRH moduli is made with the use of a high-speed computer by calculating the mean velocity of sound in crystals and comparing this result with the corresponding quantity calculated from the actual polycrystalline elastic moduli. The general agreement is observed.

## 1. INTRODUCTION

The Voigt-Reuss-Hill approximation is an averaging scheme by which anisotropic single-crystal elastic constants can be converted into isotropic polycrystalline elastic moduli. The averaging scheme is explicitly based on three independent theoretical contributions due to Voigt,<sup>1</sup> Reuss,<sup>2</sup> and Hill<sup>3</sup>; thus it was named the Voigt-Reuss-Hill (VRH) approximation.4,5 The VRH approximation is simple and easily tractable in theory. For a single-phase crystalline aggregate made of crystals that are slightly anisotropic, the approximation gives the realistic values of isotropic elastic moduli. However, for aggregates containing crystals of highly anisotropic crystals, a question arises whether or not this approximation gives still the realistic estimates of the polycrystalline elastic properties.

The purpose of this paper is to study the VRH approximation for highly anisotropic cubic crystals and to examine to what extent this approximation is useful for calculating the polycrystalline isotropic moduli from the corresponding single-crystal data. The materials chosen for the present work are MgO, CaF2, B-ZnS, ZnSe, and CdTe for which the well-characterized polycrystalline specimens were available. The polycrystalline  $\beta$ -ZnS, ZnSe, and CdTe are of particular interest in the present work since they are aggregate examples of highly anisotropic crystals having the same structure, and thus the measurements of the isotropic elastic moduli on these materials provide important empirical data that test the validity of the VRH approximation for high elastic anisotropy.

### 2. EXPERIMENTAL PROCEDURE

### 2.1. Specimens

The specimens used in the present work are vacuum hot-pressed polycrystalline aggregates and they are virtually free from porosity. These materials are known under a trade name Irtran, available from the Eastman Kodak Company. The specimens were about 0.635 by 0.953 by 7.620 cm in dimension, sufficiently large enough to make accurate resonance measurements as well as measurements of sound velocities. The faces of each specimen were polished to give a rectangular prism whose sides and ends are square and parallel to  $\pm 0.001$  cm. Using ultrasonic velocity measurements, each specimen was checked for the elastic isotropy by rotating the transducer crystals on the specimen surface. All the specimens were found to be isotropic for both the longitudinal and shear waves. In the following, a brief description of these specimens is made under each material headings.

### A. Polycrystalline MgO

One specimen designated hereafter as IR-MgO is a typical Irtran No. 5 material. The measured density was 3.5819 ( $\pm 0.0009$ ) g/cm<sup>3</sup> at 298°K and it should be compared with the x-ray density of 3.581 in the same unit. The chemical purity of IR-MgO was 99.98% MgO. Minor constituents contained in the specimen were as follows: Si 80, Ca 30, Al 25, Fe 25, Cu 10, Sn 5, Ba less than 3, B less than 10, Ni 5, Na 3, Li 2, Cr 4, K and Sr each less than 1 ppm. The specimen was highly translucent in the visible region, and it had an optical reflectance better than 60% in the 14 to 26  $\mu$ range. The measured reststrahlen lattice wavelength was 25.3  $(\pm 0.2) \mu.^{6}$ 

### B. Polycrystalline $CaF_2$

One specimen designated under IR-CaF<sub>2</sub> is a typical of Irtran No. 3 material. The measured density for the

2535

ons leading

e streamer e diagram t streamer  $10^7$  cm/sec. secondary w appears thtly\_later hode. This d negative indicated. nduced by avalanche o cathode gure. This

reakdown 5 kV

> ENSITY OF STREAMER

ce of events idary at the thode a fast irected posie light from been drawn mistakenly

## ig positive

narks the pressures this, light ocess may supply is resulting low-pres-

encourage-. B. Loeb, go to the ed me the rtment of the Office is project.

<sup>1</sup>W. Voigt, Lehrbuch der Kristallphysik (B. B. Teubner,

<sup>&</sup>lt;sup>1</sup>W. Vojet, Lehrbuch der Kristallphysik (B. B. Teubner, Leipzig, 1928), p. 739.
<sup>2</sup>A. Reuss, Z. Angew. Math. Mech. 9, 49 (1929).
<sup>3</sup>R. Hill, Proc. Phys. Soc. (London) 65, 349 (1952).
<sup>4</sup>D. H. Chung, Phil. Mag. 8, [89] 833 (1963).
<sup>4</sup>O. L. Anderson, J. Phys. Chem. Solids 24, 909 (1963). The VRHG (Voigt-Reuss-Hill-Gilvarry) approximation so called by Anderson is exactly the same as the VRH (Voigt-Reuss-Hill) approximation. See, for example, *Physical Acoustics*, edited by W. P. Mason, (Academic Press, New York, 1965), Vol. III-B, Chan, 2. Chap. 2.

<sup>6</sup> Kodak Irtran 5 Material (Eastman Kodak Company, Rochester, New York, 1963).

			Elastic modulusª		
Specimen	(g/cm <sup>3</sup> )	Young's	Şhear	Longitudinal	Method of measurements
IR-MgO	3.5819(±0.0009)	$30.72(\pm 0.12)$	$12.93(\pm 0.10)$ $12.90(\pm 0.03)$	 33.83(±0.03)	Resonance Pulse superposition
IR-CaF2	3.1792(±0.0008)	10.75(±0.09)	$4.07(\pm 0.07)$ $4.11(\pm 0.03)$	 14.87(±0.03)	Resonance Pulse superposition
IR-ZnS	4.0791(±0.0009)	8.38(±0.09)	$3.18(\pm 0.07)$ $3.17(\pm 0.03)$	11.98(±0.03)	Resonance Pulse superposition
IR-ZnSe	$5.2664(\pm 0.0009)$	7.46(±0.10)	$2.81(\pm 0.07)$ $2.89(\pm 0.03)$	9.83(±0.03)	Resonance Pulse superposition
IR-CdTe	5.8520(±0.0008)	••••	$1.40(\pm 0.05)$ $1.38(\pm 0.03)$	6.07(±0.06) 6.08(±0.03)	Phase comparison Pulse superposition

TABLE I. Measured isotropic elastic moduli of polycrystalline MgO, CaF2, β-ZnS, ZnSe, and CdTe.

<sup>n</sup> All values are at 298°K. All moduli are in units of 10<sup>11</sup> dyn/cm<sup>2</sup>.

IR-CaF<sub>2</sub> was 3.1792 (±0.0008) g/cm<sup>3</sup> at 298°K, and this value compares well with an x-ray density of 3.179 g/cm<sup>3</sup> at 298°K. The chemical purity of the specimen was 99.92% CaF<sub>2</sub>, and a spectrochemical analysis showed the following impurities: Sr 700, Na 50, Mg 20, Si 3, and Mn 2 ppm with trances of Al, Ba, K, and Li. The IR-CaF<sub>2</sub> was highly translucent in the visible region as in the case of IR-MgO and it had the optical properties corresponding to a single-crystal CaF2.

## C. Polycrystalline $\beta$ -ZnS

The  $\beta$ -ZnS specimen used in the present program is a typical of Irtran No. 2 material. The measured density was 4.0791 ( $\pm 0.0009$ ) g/cm<sup>3</sup> at 298°K, and this should be compared with a x-ray density of 4.088 g/cm<sup>3</sup> at the same temperature. The specimen has been referred to as IR-ZnS in the text, and it has a chemical purity of 99.997% ZnS. Among the impurities detected were Si 1 and Pb 1 ppm with traces of Cu, Fe, and Mg. X-ray diffraction patterns indicate the IR-ZnS was composed primarily of  $\beta$ -ZnS (i.e., sphalerite).

### D. Polycrystalline ZnSe

One specimen of polycrystalline ZnSe used in the present program is a typical Irtran No. 4 material, and this has been designated as IR-ZnSe. The IR-ZnSe had the bulk density of 5.2664 ( $\pm 0.0009$ ) g/cm<sup>3</sup> at 298°K and this value compares well with 5.267 gm/cm3, the x-ray density at the same temperature. This specimen has a chemical purity of 99.995% ZnSe, with minor constituents including Mg 5, Cu 3, B 10, and Al 2 ppm and traces of Cr and Ni. The IR-ZnSe was translucent in the visible region and had the optical properties similar to single-crystal ZnSe.

## E. Polycrystalline CdTe

One specimen referred hereafter to as IR-CdTe is a typical of Irtran No. 6 material. The measured density was 5.8520 (±0.0008) g/cm<sup>3</sup> at 298°K and this may be compared with 5.854 gm/cm<sup>3</sup> calculated from the lattice constant<sup>7</sup> of 6.5815 Å at 298°K. The chemical purity of the specimen was 99.98% CdTe and the specimen contained the following impurities: B 10, Si 10, Mg 3 ppm and traces of Al, Ag, and Cu.

### 2.2. Measurements of Isotropic Elastic Moduli

The present work utilizes primarily a modified Förster-type resonance method in the kilocycle range<sup>8</sup> to determine the isotropic shear modulus and then Young's modulus of a bar-shaped polycrystalline specimen. Two ultrasonic methods9 (phase-comparison and pulse-superposition techniques due to McSkimin) often used in single-crystal measurements are also used, as complementary methods, for determining the elastic parameters of polycrystalline solids under investigation. Since a detailed description on all of these methods are found in the literature, this description is not reproduced here.

#### 3. EXPERIMENTAL RESULTS

Table I lists the measured elastic moduli for all the polycrystalline specimens considered in the present work. For a given material, two sets of isotropic elastic moduli are entered. One set is the result obtained from the resonance method in the audio-frequency range and

2536

Mat reference

> MgO MgO

MgO MgO MgO MgO CaF<sub>2</sub> CaF2 CaF2 CaF2 CaF2 B-ZnS B-ZnS B-ZnS B-ZnS B-ZnS B-ZnS ZnSe

<sup>a</sup> Values of the origi represent b 36D1: c 61S1: d 63C1: 46,452 (190 ° 65B1: 1 f 65C1: # 28V1: N h 58S1: 1

<sup>i</sup> 60H1: 1

CdTe

CdTe

the othe in the dampin were no ured by parison from th moduli, the she indicate determi the gen the ultra

4. SINC

For n than one in the lit

 <sup>&</sup>lt;sup>7</sup> P. W. Davis and T. S. Shilliday, Phys. Rev. 118, 1020 (1960).
 <sup>8</sup> S. Spinner and W. E. Tefft, Proc. ASTM 61, 1221 (1961).
 <sup>9</sup> H. J. McSkimin, *Physical Acoustics* W. P. Mason, Ed. (Academic Press Inc., New York, 1964), Vol. I-A, Chap. 4.

### VOIGT-REUSS-HILL APPROXIMATION

TABLE II. Literature values of single-crystal elastic constants of MgO, CaF2, β-ZnS, ZnSe, and CdTe.ª

r	eference temperature (°K)	Density (g/cm³)	S <sub>11</sub> ()	− <i>s</i> <sub>12</sub> ×10 <sup>13</sup> cm²/dyr	S44	$^{C_{11}}$ (×	10 <sup>-11</sup> dyn/c	C44 m <sup>2</sup> )	A	Reference
-	MgO (300)	3.576	4.024	0.936	6.461	28.93	8.77	15.477	1.54	36D1 <sup>b</sup>
	MgO (298)	3.580	(4.075)	(0.950)	(6.757)	28,60	8.70	14.80	1.49	55B1*
	MgO (293)	3.598	3.983	1.000	6.349	30.20	10.10	15.75	1.57	61S1°
	MgO (300)	3.579	4.029	0.942	6.468	28.94	8.82	15.46	1.54	63C1d
	MgO (298)	3.579	(3.988)	(0.969)	(6.405)	29,708	9.536	15.613	1.55	65B1e
	MgO (297)	3.581	4.004	0.973	6.428	29.60	9.51	15.557	1.55	65C1f
	CaF <sub>2</sub> (300)	3.180	(6.904)	(1.479)	(29.586)	16.4	4.47	3.38	0.57	28V1#
	CaF <sub>2</sub> (298)	3.180	(7.097)	(1.660)	(28.814)	16.44	5.02	3.47	0.61	55B1s
	$CaF_{2}(0)$	3.180	(6.818)	(1.515)	(25.000)	16.8	4.8	4.0	0.67	58S1h
	CaF <sub>2</sub> (300)	3.1795	(7.241)	(1.768)	(29.674)	16.4	5.3	3.37	0.61	60H11
	CaF <sub>2</sub> (293)	3.1804	(6.901)	(1.463)	(29.481)	16.357	4.401	3.392	0.57	63H1 <sup>i</sup>
	β-ZnS (298)	4.102	(19.429)	(7.309)	(22.936)	9.42	5.68	4.36	2.33	18V1 <sup>k</sup>
	β-ZnS (298)	4.102	(19.995)	(8.016)	(24.272)	10.79	7.22	4.12	2.31	44B11
	β-ZnS (298)	4.102	(20.497)	(8.075)	(29.412)	10.0	6.5	3.4	1.94	51P1m
	β-ZnS (302)	4.079	(18.817)	(7.090)	(22.173)	9.76	5.90	4.51	2.34	63E1n
	β-ZnS (298)	4.088	(18.381)	(7.065)	(21.678)	10.46	6.53	4.613	2.35	63B1°
	β-ZnS (293)	4.083	(18.706)	(7.201)	(21.645)	10.32	6.46	4.62	2.39	63Z1ª
	ZnSe (298)	5.262	(22.570)	(8.486)	(22.676)	8.10	4.88	4.41	2.74	63B1°
	CdTe (298)	5.854	(42.542)	(17.338)	(50.151)	5.351	3.681	1.994	2.39	62M1r
	CdTe (77)	5.860	(38.296)	(15.758)	(51.020)	6.15	4.30	1.96	2.12	63B1º

 $^{\rm a}$  Values in parentheses are the present writers' computation based on work of the original authors. The number of the significant figures does not necessarily

represent the accuracy of the values.

measurements

rposition

rposition

position

position parison position

-CdTe is a red density d this may

d from the

he chemical

d the speci-

10, Si 10,

Moduli

1 modified ycle range<sup>8</sup> and then

lline speci-

arison and

McSkimin)

e also used.

the elastic

restigation.

ethods are

not repro-

for all the

ie present

pic elastic

lined from

range and

020 (1960). 221 (1961).

i, Ed. (Aca-

<sup>b</sup> 36D1; M. A. Durand, Phys. Rev. 59, 449 (1936).
 <sup>c</sup> 61S1; C. Susse, J. Res. CNSR (Paris), 54, 23 (1961).

<sup>d</sup> 63C1: D. H. Chung, J. J. Swica, and W. B. Crandall, J. Am. Ceram. Soc.

46, 452 (1963).

° 65B1: E. Hal Bogardus, J. Appl. Phys. 36, 2504 (1965).

<sup>f</sup> 65C1: D. II. Chung (unpublished work, 1965).

# 28V1: W. Voigt Lehrbuch der Kristallphysik, (Teubner, Berlin, 1928), p. 744.

h 58S1: R. Srinivasan, Proc. Phys. Soc. (London), 72, 556 (1958).

<sup>1</sup> 60II1: D. R. Huffman and M. H. Norwood, Phys. Rev. 117, 709 (1960).

the other from McSkimin's pulse-superposition method in the ultrasonic frequency range. Because of high damping, the resonance measurements on IR-CdTe were not made but instead sound velocities were measured by both the pulse-superposition and phase-comparison methods. The primary constants resulting from the resonance method are the shear and Young's moduli, whereas those from the ultrasonic methods are the shear and longitudinal moduli. In each case, the combined experimental errors in the modulus are indicated. It is noted here that the elastic moduli determined from the resonance method are always in the general agreement with the ones determined from the ultrasonic methods.

## 4. SINGLE-CRYSTAL ELASTIC CONSTANTS AND CALCULATIONS OF THE VOIGT-REUSS-HILL (VRH) MODULI

For most materials considered here, there are more than one set of single-crystal elastic constants reported in the literature and in some cases they differ in values <sup>1</sup>63111: S. Haussuhl, Phys. Status Solidi 3, 1072 (1963).

k 18V1: W. Voigt, Göttinger Nachr. 424 (1918).

<sup>1</sup>44B1: S. Bhagavantam and D. Suryanarayana, Proc. Ind. Acad. Sci., A20, 304 (1944).

<sup>m</sup> 51P1: E. Prince and W. A. Wooster, Acta Cryst. 4, 191 (1951).

<sup>n</sup> 63E1: N. G. Einspruch and R. J. Manning, J. Acoust. Soc. Am. 35, 215

(1963). <sup>a</sup> 63B1: D. Berlincourt, H. Jaffe, and L. R. Shiozawa, Phys. Rev. 129, 1009 (1963).

<sup>q</sup> 63Z1: A. Zarembovitch, J. Phys. (Paris) 24, 1097 (1963).

<sup>r</sup> 62M1: H. J. McSkimin and D. G. Thomas, J. Appl. Phys. 33, 56 (1962).

<sup>8</sup> 55B1: S. Bhagavantam, Pro. Ind. Ind. Acad. Sci., A41,: 78 (1965).

from one author's result to another. This situation presents a problem of selecting the elastic constants realistic of the solid. For this reason, all the known values of elastic constants are quoted in Table II as they found in the literature. In each case, entries have been made for the reference temperature after the material heading and also for the density of the specimen used by the original author, the elastic constants, the elastic anisotropy factor, and the source of this information. Using these single-crystal data, the isotropic VRH moduli are calculated according to the relations

and

and

$$K_{\rm VRII} = K_V = K_R = K^* = c_{11} - 2C/3 \tag{1}$$

$$G_{\rm VRH} = G^* = (G_V + G_R)/2,$$
 (2)

where  $C = (c_{11} - c_{12})$ . The limiting moduli  $G_V$  and  $G_R$  are given by

$$G_V = C/5 + 3c_{44}/5,$$
 (3)

$$G_R = 5Cc_{44}/(4c_{44} + 3C), \qquad (4)$$

Material and reference temperature (°K)	Reference to c <sub>ij</sub> <sup>b</sup>	Kvrm	$G_{\rm VRH}$	$E_{\rm VRH}$	$G_V - G_R$	$E_V - E_R$
MgO (300) MgO (298) MgO (293) MgO (300) MgO (298) MgO (297)	36D1 55B1 61S1 63C1 65B1 65C1	$\begin{array}{c} 15.490 \\ 15.333 \\ 16.800 \\ 15.530 \\ 16.260 \\ 16.205 \end{array}$	$\begin{array}{c} 13.033\\ 12.623\\ 13.154\\ 13.013\\ 13.104\\ 13.056\end{array}$	30.532 29.712 31.291 30.513 30.985 30.873	$\begin{array}{c} 0.571 \\ 0.475 \\ 0.632 \\ 0.573 \\ 0.596 \\ 0.595 \end{array}$	1.045 0.877 1.194 1.051 1.112 1.109
$\begin{array}{c} CaF_2 (300) \\ CaF_2 (298) \\ CaF_2 (0) \\ CaF_2 (300) \\ CaF_2 (293) \end{array}$	28V1 55B1 58S1 60H1 63H1	8.447 8.827 8.800 9.000 8.386	$\begin{array}{c} 4.251 \\ 4.241 \\ 4.708 \\ 4.120 \\ 4.264 \end{array}$	10.920 10.965 11.985 10.723 10.936	$\begin{array}{c} 0.325 \\ 0.250 \\ 0.185 \\ 0.244 \\ 0.324 \end{array}$	0.716 0.557 0.399 0.550 0.712
β-ZnS (298) β-ZnS (298) β-ZnS (298) β-ZnS (302) β-ZnS (298) β-ZnS (293)	18V1 44B1 51P1 63E1 63B1 63Z1	6.930 8.410 7.667 7.187 7.840 7.747	$\begin{array}{c} 3.112 \\ 2.945 \\ 2.604 \\ 3.208 \\ 3.276 \\ 3.255 \end{array}$	8.114 7.908 7.017 8.372 8.619 8.558	0.520 0.481 0.271 0.539 0.556 0.578	$\begin{array}{c} 1.180\\ 1.157\\ 0.656\\ 1.226\\ 1.280\\ 1.333 \end{array}$
ZnSe (298)	63B1	5.953	2.945	7.573	0.689	1.524
CdTe (298) CdTe (77)	62M1 63B1	$4.238 \\ 4.917$	$1.406 \\ 1.450$	3.796 3.959	0.248 0.192	$0.604 \\ 0.477$

TABLE III. The Voigt-Reuss-Hill moduli of MgO, CaF2, β-ZnS, ZnSe, and CdTe.ª

<sup>a</sup> All moduli are in units of 10<sup>11</sup> dyn/cm<sup>2</sup>.

<sup>b</sup> See Table II for the complete references.

respectively. Knowing the bulk modulus  $K^*$  and the shear modulus  $G^*$ , one can calculate Young's modulus  $E^*$ , the longitudinal modulus  $L^*$ , and Lame's constant  $\lambda^*$  as well as the compressibility  $\chi^*$  and Poisson's ratio  $\mu^*$  from the well-known relations<sup>10</sup> of the isotropic elasticity.

The result of these calculations for the isotropic bulk, shear, and Young's moduli are tabulated in Table III for all the single-crystal data listed in Table II. Also entered are the differences in the limiting Voigt and Reuss moduli since they are related to the actual magnitude of the elastic anisotropy possessed by crystals.<sup>11</sup> Table IV compares selected values of the measured and calculated isotropic moduli from Table I and Table III, respectively.

### 5. DISCUSSION

It is apparent from Table IV that the measured polycrystalline elastic moduli are in good agreement with the isotropic VRH moduli calculated from the corresponding single-crystal elastic constants. The differences observed between the measured and calculated values are in most cases within the scatters in the calculated VRH moduli resulting from the differences in the single-crystal elastic constants of one author to another (see Table III). This indicates the success of

TABLE IV. Comparison between the measured and calculated isotropic elastic moduli of MgO, CaF2, β-ZnS, ZnSe, and CdTe.

			Ε	lastic modulusª		
		Material and references <sup>b</sup>	Shear	Young's	Bulk	
	MgO	Single-crystal (65C1) polycrystalline	$13.056 \\ 12.93(\pm 0.10)$	30.873 30.72(±0.12)	16.205 16.41	
	$\operatorname{CaF}_2$	Single-crystal (60H1) polycrystalline	4.120 4.07(±0.07)	$10.723 \\ 10.75(\pm 0.09)$	9.000 9.98	
	$\beta$ -ZnS	Single-crystal (63E1) polycrystalline	3.208 3.18(±0,07)	8.372 8.38(±0.09)	7.187 7.66	
	ZnSe	Single-crystal (63B1) polycrystalline	2.945 $2.88(\pm 0.07)$	7.573 $7.46(\pm 0.10)$	5.953 6.67	
	CdTe	Single-crystal (62M1) polycrystalline	$1.406 \\ 1.38(\pm 0.03)$	3.796 3.73	4.238 4.25	

<sup>a</sup> All moduli are in units of 10<sup>11</sup> dyn/cm<sup>2</sup>.

<sup>10</sup> F. Birch, J. Geophys. Res. 65, 3855 (1960); Table 4.
 <sup>11</sup> D. H. Chung and W. R. Buessem, J. Appl. Phys. (in press).

<sup>b</sup> See Table II for the complete references.

where eigenv

the V crystal To approx ratio o moduli Figs. 1 here is A\*(i)where and R elastic worthy the spr program (Gmens/ deviati elastic tion ca

Ma

MgO

CaF2

B-ZnS

CdTe

ZnSe

<sup>a</sup> All v <sup>b</sup> See

The tr anisotr

like Li the goo possess  $A^* < 10$ accura and th in the l To p we tak mean compa deduce mean of the second the VRH approximation, even for the anisotropic crystals like CdTe and ZnSe.

To examine a possible dependence of the VRH approximation on elastic anisotropy, we plotted the ratio of the measured modulus to the calculated VRH modulus as a function of percent elastic anisotropy in Figs. 1 and 2. The percent elastic anisotropy<sup>11</sup> referred here is

$$A^*(\inf \%) = [3(A-1)^2/3(A-1)^2 + 25A] \times 100, \quad (5)$$

where  $A = 2c_{44}/(c_{11}-c_{12})$ . Note that the limiting Voigt and Reuss moduli result in the wider spread as the elastic anisotropy of crystal becomes large. It is noteworthy, however, that the measured moduli lie within the spread for every crystals considered in the present program. As seen in Fig. 2, the ratio of shear moduli  $(G_{\text{meas}}/G_{\text{VRH}})$  is smaller than unity in all cases and the deviation of this ratio from unity becomes large as the elastic anisotropy of crystal increases. Similar observation can be made also for the case of Young's modulus.

TABLE V. Mean velocity of sound for MgO, CaF<sub>2</sub>,  $\beta$ -ZnS, CdTe, and ZnSe.<sup>a</sup>

M	aterials and reference <sup>b</sup>		$\begin{bmatrix} v_m \\ \text{Eq. (6)} \end{bmatrix}$	$\begin{bmatrix} v_m^* \\ [Eq. (7)] \end{bmatrix}$
MgO	Single-crystal (65C1) polycrystalline	2.28	6.617	6.654
$CaF_2$	Single-crystal (60H1) polycrystalline	2.96	4.001	4.022
β-ZnS	Single-crystal (63E1) polycrystalline	8.49	3.122	3.135 3.127
CdTe	Single-crystal (62M1) polycrystalline	8.83	1.712	$1.743 \\ 1.727$
ZnSe	Single-crystal (63B1) polycrystalline	11.70	2.406	$2.637 \\ 2.614$

<sup>a</sup> All values of the velocity are in units of 10<sup>5</sup> cm/sec.

<sup>b</sup> See Table II for the complete references.

The trend of this deviation with increasing elastic anisotropy suggests that, for highly anisotropic crystals like Li and RbI, the VRH approximation may not be the good procedure to follow. But, for the cubic crystals possessing low or moderate elastic anisotropics (i.e.,  $A^* < 10\%$ ), the VRH approximation is believed to be accurate in giving the probable isotropic elastic moduli and these VRH moduli are as good as ones we measure in the laboratory.

To provide an additional support to this conclusion, we take a numerical approach in which we calculate the mean velocity of sound in a given crystal and then compare this result with the corresponding quantities deduced from the Debye continuum relation. The mean velocity of sound in an anisotropic crystal is

$$v_m = \left[\frac{1}{3} \sum_{j=1}^{3} \int_V \left(\frac{1}{v_j^3}\right) \frac{d\Omega}{4\pi}\right]^{-1/3}, \ (j=1,\,2,\,3), \qquad (6)$$

where  $v_j$  represent three sound velocities that are the eigenvalues of the Christoffel equation involving the



FIG. 1. Comparison between calculated and measured Young's moduli as a function of elastic anisotropy.

single-crystal elastic constants and  $d\Omega$  is the element of a solid angle, i.e.,  $d\Omega = \sin\theta d\theta d\phi$ . Since the integration of Eq. (6) is impractical to perform analytically, the integration is evaluated numerically as a procedure outlined by Alers.<sup>12</sup> Using the single-crystal elastic constants for the individual materials considered, values of the mean velocity of sound have been calculated by





<sup>12</sup> G. A. Alers, *Physical Acoustics*, W. P. Mason, Ed. (Academic Press Inc., New York, 1965), Vol. III-B, Chap. 1.

and Table III,

ER

the measured od agreement ted from the nstants. The ed and calcucatters in the he differences one author to he success of

#### e, and CdTe.

use of TBM-7074 computer. The computed values of  $v_m$  are tabulated in Table V and these are compared with values calculated from (i) the VRH moduli and (ii) the measured polycrystalline elastic moduli presented earlier in Table I. Since the polycrystalline elastic moduli are isotropic, such a calculation of the mean velocity of sound from the polycrystalline data is simply done by the use of a Debye expression<sup>13</sup>

$$v_m^* = \begin{bmatrix} \frac{1}{3} (1/v_l^{*3} + 2/v_l^{*3}) \end{bmatrix}^{-1/3}, \tag{7}$$

where  $v_l^*$  and  $v_l^*$  are the isotropic longitudinal and transverse velocities of sound, respectively, and they are defined by the isotropic longitudinal and shear moduli in the usual way.

<sup>13</sup> See, for example, T. H. K. Barron, Phil. Mag. 7, [46] 720 (1955) and also Ann. Phys. 1, 77 (1957).

It is evident from Table V that the values of the mean velocity of sound calculated from the singlecrystal data agree well with that obtained from the polycrystalline data. In other words, the values of  $v_m^*$ calculated from Eq. (7) using both the VRH moduli and the actual polycrystalline moduli find in the general agreement with the  $v_m$  calculated from Eq. (6). The kind of the agreement observed here supports the earlier conclusion that the VRH approximation gives realistic values of the polycrystalline elastic moduli in terms of the corresponding single-crystal properties.

### ACKNOWLEDGMENTS

The writers acknowledge Dr. Edward Carnall, Jr. for providing the Irtran samples. This work was supported by U.S. Office of Naval Research.

## JOURNAL OF APPLIED PHYSICS

VOLUME 38, NUMBER 6

MAY 1967

the

lase

esti

the

T

and

whe the exa

for

liv =

pur

77 :

COL

ap

COL

ph

aci

di

en

fo

of

re

# Behavior of Saturable-Absorber Giant-Pulse Lasers in the Limit of Large Absorber Cross Section

L. E. ERICKSON AND A. SZABO

Radio & Electrical Engineering Division, National Research Council, Ottawa, Canada

(Received 9 January 1967)

Using a rate-equation model, it is shown that the behavior of the saturable-absorber giant-pulse (SAGP) laser can be adequately described in terms of two parameters for values of the ratio of absorber to laser absorption cross section  $\sigma > 200$ :  $n_{ai}'$  the normalized initial inversion and  $\sigma \tau_s$ , where  $\tau_s$  is the normalized absorber relaxation time. In the general case, specification of  $n_{ai}', \sigma$ , and  $\tau_s$  is required. Theoretical curves of the giant-pulse output power, energy, and rise- and falltimes are presented. The results are applicable in particular to SAGP lasers employing organic-dye absorbers.

### INTRODUCTION

Recently, the saturable-absorber giant-pulse (SAGP) laser has been the subject of intensive study.<sup>1-16</sup> SAGP

<sup>1</sup> P. P. Sorokin, I. J. Luzzi, J. R. Lankard, and G. D. Pettit, IBM J. Res. Develop. 8, 182 (1964). <sup>2</sup> M. T. Melamed and C. Hirayama, Appl. Phys. Letters 6, 43

(1965)

<sup>3</sup> E. Snitzer and R. Woodcock, IEEE J. Quantum Electron.

QE-2, 627 (1966). <sup>4</sup> H. W. Gandy, R. J. Ginther, and J. F. Weller, Appl. Phys. Letters 9, 277 (1966).

<sup>6</sup> S. Yoshikawa and Y. Matumura, IEEE J. Quantum Electron, QE-2, xlviii (1966).

 <sup>6</sup> A. Szabo and R. A. Stein, J. Appl. Phys. 36, 1562 (1965).
 <sup>7</sup> L. E. Erickson and A. Szabo, J. Appl. Phys. 37, 4953 (1966).
 <sup>8</sup> R. McLeary and P. W. Bowe, Appl. Phys. Letters 8, 116 (1966).

 <sup>9</sup> W. R. Sooy, Appl. Phys. Letters 7, 36 (1965).
 <sup>10</sup> H. W. Mocker and R. J. Collins, Appl. Phys. Letters 7, 270 (1965).

<sup>11</sup> E. R. Peressini and W. R. Sooy, J. Opt. Soc. Am. 56, 1433 (1966).

12 A. J. DeMaria, D. A. Stetser, and H. Heynau, Appl. Phys. Letters 8, 174 (1966).
<sup>13</sup> B. H. Soffer, J. Appl. Phys. 35, 2551 (1964).
<sup>14</sup> D. Roess and G. Zeidler, Appl. Phys. Letters 8, 10 (1966).
<sup>15</sup> F. J. McClung and D. Weiner, IEEE J. Quantum Electron.

QE-1, 94 (1965).

16 V. Dancu, C. A. Sacchi, and O. Svelto, IEEE J. Quantum Electron. QE-2, 290 (1966).

lasers might be classed into two types, depending on the nature and distribution of absorber centers in the laser cavity. The more common device uses an organicdye absorber, which is physically separated from the amplifying medium. Such dyes have absorption cross sections which are typically 103 to 105 times larger than that of the laser centers. In the other type of SAGP laser, the absorber is uniformly distributed throughout the amplifying medium, e.g., Nd3+ glass co-doped with UO22+,2 color centers in Nd3+ glass3 and Ho3+ glass codoped with Fe2+.4 There have also been indications of self Q-switching in ruby co-doped with Ti and Fe.5 The absorber parameters in the latter systems are, as yet, largely undetermined.

In earlier publications<sup>6,7</sup> a theory of SAGP lasers was formulated in terms of three parameters,  $n'_{ai}$  the normalized inversion prior to Q switching,  $\sigma$  the ratio of absorber to laser cross section, and  $\tau_s$  the absorber lifetime normalized to the cavity photon lifetime. It is the purpose of this paper to show that for sufficiently large  $\sigma$ , the SAGP laser behavior can be adequately described by only two parameters,  $n'_{ai}$  and the product  $\sigma \tau_s$ . The range of validity of this description is examined in detail and is shown to be a good approximation for

2540