Determination of Third-Order Elastic Constants from Stress-Shifted Resonance Frequencies, Observed by Diffraction of Light. Examples: Aluminium Alums of K, NH₄, Cs and CH₃NH₃

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Resonance frequencies of thick plane-parallel plates are easily detected by diffraction of light with an accuracy better than 1 p.p.m. The method, which has been earlier employed for the determination of thermoelastic properties, proved to be sufficiently sensitive also for the measurement of stress-induced shifts of resonance frequencies, and is therefore suited for the determination of third-order elastic constants. The technique is easier to handle and much less expensive than other methods in use today. The complete sets of third-order elastic constants for $KAl(SO_4)_2.12H_2O$, $NH_4Al(SO_4)_2.12H_2O$, $CsAl(SO_4)_2.12H_2O$ and $CH_3NH_3Al(SO_4)_2.12H_2O$ have been measured. All constants possess negative values. These alums exhibit a behaviour similar to that of the structurally related caesium halides, with the exception of ammonium alum which shows anomalously small constants.

Introduction

The nonlinear elastic behaviour of solids, represented by higher-order elasticity tensors, is directly related to all anharmonic properties such as thermal expansion, thermoelasticity, and attenuation processes of phonon propagation. In order to establish general equations of state covering higher stress and temperature ranges a knowledge of the third-order elastic constants (TOEC) is required.

A survey of the experimental procedures for measuring TOEC has been given by Wallace (1970). Highly sensitive pulse-echo and pulse-transmission interference methods have been developed by Forgacs (1960, 'sing-around') and McSkimin (1961, 'pulse superposition').

In this paper we report results obtained with the much simpler and less expensive method of measuring resonance frequencies of thick plane-parallel plates. The same technique has been successfully used for the recent determination of thermoelastic constants of numerous crystals by one of us (Haussühl, 1960, 1961, 1963). The accuracy achieved during routine work proved to be sufficient for the evaluation of the complete set of TOEC of crystals. For testing the method we measured the aluminium alums of K, NH₄, Cs and CH₃NH₃ which all belong to symmetry group *m*3. Specimens of high optical quality were available from earlier investigations (Haussühl, 1961). No crystals of this symmetry group have been investigated so far.

Experimental

Basic equations for the determination of nonlinear acoustic properties of crystals have been derived by Seeger & Buck (1960) and by Thurston & Brugger (1964). A thorough review article has been published by Wallace (1970). In our paper we use the notations introduced there. The observable magnitudes are the second-order elastic constants (SOEC) at zero stress, c_{ijkl}, the zero-stress density ρ_0, and the stress derivatives of ultrasonic velocities v of plane elastic waves, with propagation unit vector **k** and displacement unit vector **w**. The following fundamental equation combines these observed quantities with the unknown TOEC c_{ijklmn}:

$$\partial(\rho_0 W^2)/\partial\sigma_{pq} = k_p k_q + 2\rho_o W^2 s^T_{ijpq} w_i w_j$$
$$+ c_{ijklmn} s^T_{mnpq} k_j k_l w_i w_k.$$

All quantities are taken at zero stress. (The bars used by Wallace to denote zero-stress conditions are omitted here.) W is the so-called 'natural velocity' defined by $W = (l_0/l_1)v$, where l_0 and l_1 denote the length of the specimen in the direction of the propagation vector without and with applied stress respectively. σ_{pq} are the components of the applied static stress tensor. The second and third-order elastic constants are defined in the sense of Brugger (1964) as

$$c_{ijkl} = \rho_0(\partial^2 U/\partial\eta_{ij}\partial\eta_{kl})$$

and

$$c_{ijklmn} = \rho_0(\partial^3 U/\partial\eta_{ij}\partial\eta_{kl}\partial\eta_{mn}).$$

Table 1. *Arrangements of measurement and equations for the evaluation of TOEC for symmetry group m3*

In Table 1 each equation is read horizontally, e.g. for propagation mode No. 1: $c_{111} + c_{112} + c_{113} = B_1$, where B_1 is directly related to experimental data (Table 2). z is the direction of the light beam; $a_1 = s^T_{11}/s^T_{12}$, $a_2 = (s^T_{11} + 3s^T_{12})/2(s^T_{11} + s^T_{12})$, $a_3 = s^T_{44}/(s^T_{11} + s^T_{12})$, $a_4 = (s^T_{11} + s^T_{12})/2s^T_{12}$; s^T_{ij}: isothermal second-order elastic compliance.

No.	Wave type*	Direction of stress	k	w	z	c_{111}	c_{112}	c_{113}	c_{123}	c_{144}	c_{155}	c_{166}	c_{456}	B_i
1	l	hydrostatic	[100]	[100]	[010]	1	1	1	0	0	0	0	0	B_1
2	t	hydrostatic	[100]	[010]	[001]	0	0	0	0	1	1	1	0	B_2
3	l	hydrostatic	[111]/$\sqrt{3}$	[111]/$\sqrt{3}$	[1$\bar{1}$0]	1	3	3	2	4	4	4	0	B_3
4	t	hydrostatic	[111]/$\sqrt{3}$	[1$\bar{1}$0]/$\sqrt{2}$	[11$\bar{2}$]	1	0	0	−1	1	1	1	0	B_4
5	l	hydrostatic	[110]/$\sqrt{2}$	[110]/$\sqrt{2}$	[001]	1	2	2	1	2	2	2	0	B_5
6	t	hydrostatic	[110]/$\sqrt{2}$	[001]	[1$\bar{1}$0]	0	0	0	0	1	1	1	0	B_6
7	t	hydrostatic	[110]/$\sqrt{2}$	[1$\bar{1}$0]/$\sqrt{2}$	[001]	1	0	0	−1	0	0	0	0	B_7
8	l	[100]	[100]	[100]	[001]	1	$1/a_1$	$1/a_1$	0	0	0	0	0	B_8
9	t	[100]	[100]	[001]	[010]	0	0	0	0	1	a_1	1	0	B_9
10	t	[100]	[100]	[010]	[001]	0	0	0	0	1	1	a_1	0	B_{10}
11	l	[100]	[010]	[010]	[001]	1	1	a_1	0	0	0	0	0	B_{11}
12	l	[100]	[001]	[001]	[010]	1	a_1	1	0	0	0	0	0	B_{12}
13	t	[100]	[001]	[100]	[010]	0	0	0	0	1	a_1	1	0	B_{13}
14	t	[100]	[010]	[100]	[001]	0	0	0	0	1	1	a_1	0	B_{14}
15	t	[001]	[110]/$\sqrt{2}$	[001]	[1$\bar{1}$0]	0	0	0	0	1	a_4	a_4	0	B_{15}
16	l	[110]/$\sqrt{2}$	[001]	[001]	[1$\bar{1}$0]	1	a_4	a_4	0	0	0	0	0	B_{16}
17	t	[110]/$\sqrt{2}$	[001]	[110]/$\sqrt{2}$	[1$\bar{1}$0]	0	0	0	0	1	a_2	a_2	a_3	B_{17}

* l = longitudinal, t = transverse.

η_{ij} are the components of the Lagrange deformation tensor at zero stress, and U is the internal energy per unit mass. s_{ijkl} are the components of the second-order elasticity s tensor (inverse c tensor). In our measurements the c_{ijklmn} are of the 'mixed' type, because the strain induced by the external stress is of static nature and the strains involved in the ultrasonic waves are dynamic. The difference between 'mixed' and adiabatic TOEC is usually smaller than the experimental error (Guinan & Ritchie, 1970). Therefore we neglected this difference.

Crystals of symmetry $m3$ possess the following three SOEC and eight TOEC (in Voigt notation: index pair $ii \rightarrow i$, index pair $ij \rightarrow 9 - i - j$ for $i \neq j$): c_{11}, c_{12}, c_{44}; c_{111}, c_{112}, c_{113}, c_{123}, c_{144}, c_{155}, c_{166}, c_{456}. For the second-order constants values of high precision are available (Haussühl, 1961). For the evaluation of the 8 TOEC we selected 17 different sets of directions of acoustic wave propagation unit vector k, acoustic wave displacement unit vector w, and light-beam direction z, as listed in Table 1. The crystals were cut into rectangular parallelepipeda with dimensions between 8 and 28 mm in accordance with the requirements of that list. The sequence of the cubic axes of the reference system is not arbitrary in crystals of symmetry $m3$. We have indexed the unique pentagonal dodecahedron which develops during natural growth in β-alums, and in a sphere growth experiment in α-alums, as {210} (Haussühl, 1961). The orientations were additionally controlled by Laue photographs. In order to obtain sharp resonance frequencies we prepared the faces plane-parallel with deviations less than 1 μm. The optical transparency was achieved by polishing and

then attaching glass plates of optical quality by the aid of an immersion fluid (paraffin oil). For the generation of ultrasonic waves, X-cut and Y-cut quartz transducers with a basic frequency of ca 5 MHz were cemented on one of the faces perpendicular to the prescribed propagation direction with paraffin oil (longitudinal waves) or synthetic DOW 276 V9 (transverse waves).

A block diagram of the experimental arrangement is given in Fig. 1. The measurements were carried out in a hydrostatic pressure cell working up to ca 1400 bar and on a device for uniaxial stresses up to ca 30 bar. The temperature was carefully kept constant. In order to reduce coupling effects between specimen and transducer, the excitation frequencies at about 14 and 27 MHz were chosen far from the resonance frequencies of the transducer.

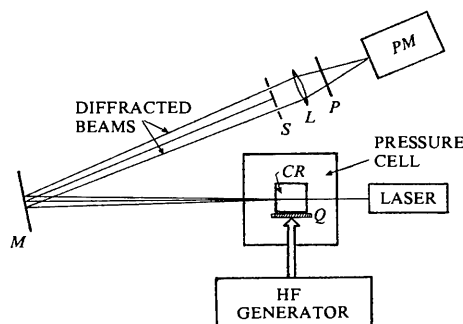In a first step we chose some resonance frequencies

Fig. 1. Block diagram of apparatus for measuring stress-induced shifts of resonance frequencies. *CR*: crystal, *Q*: quartz transducer, *M*: mirror, *S*: shield, *L*: lens, *P*: polarizer, *PM*: photomultiplier.

f_m at zero stress (m order of resonance). They are easily detected by strong, sharp light diffraction spots on a glass screen. In the case of transverse waves a polarizer, in extinction position for the primary beam, is mounted in front of the screen. By this measure the background intensity is almost fully eliminated, and the diffraction spots are excellently visible even with very weak ultrasonic amplitudes. The frequencies of the maxima of intensity are taken as resonance frequencies. Their reproducible frequency reading is within one p.p.m. Then a photomultiplier was adjusted to replace the screen and to detect the intensity maxima with a still higher accuracy. In the next step the maximum hydrostatic or uniaxial stress was applied in order to achieve a stable contact film between transducer and specimen. After unloading, the specimens were loaded step by step with increasing stresses, and then stepwise unloaded, and the resulting frequency shifts Δf were measured.

In cases No. 11 to 17 of Table 1, where uniaxial stress and propagation vector are perpendicular, the weakest influence of coupling was observed. A certain control of coupling effects is possible with the conditions $B_2 = B_6$, $B_9 = B_{13}$, and $B_{10} = B_{14}$ (Table 1). Only such measurements were accepted for the evaluation which yielded a linear relation between stress and frequency shift over the whole stress range applied. In the case of measurements No. 1 and 5 on NH_4 alum, a reproducible curvature of the slope $\partial f/\partial P$ was observed, indicating possibly the influence of higher-order nonlinear effects.

The relation between the measured quantities $\partial f/\partial\sigma_{pq}$ and the left-hand side of the basic equation is given by: $\partial(\rho_0 W^2)/\partial\sigma_{pq} = 2\rho_0 W^2(1/f)(\partial f/\partial\sigma_{pq})$ (Wallace, 1970, p. 370). With a complete set of experimental data the computation of the TOEC was carried out employing a least-squares method for the solution of the overdetermined system of linear equations (Table

1). The weight of each mode was chosen in inverse proportion to the statistical error of the quantities B_i (Table 2).

Values for auxiliary parameters (SOEC, density, coefficients of linear thermal expansion, etc.) are given by Haussühl (1961; see Table 4).

The pressure derivatives of SOEC $Pc_{ij} = \partial \ln c_{ij}/\partial P$ (P-hydrostatic pressure) were obtained by the relation

$$Pc = \partial \ln \rho/\partial P + 2\partial \ln l/\partial P + 2\partial \ln f/\partial P.$$

For cubic crystals this simplifies to:

$$Pc = 1/(c_{11} + 2c_{12}) + 2(1/f)(\partial f/\partial P).$$

Table 2. *Relations between quantities B_i (Table 1) and observable stress-induced frequency shift for symmetry group m3*

c_{ij}^S: adiabatic SOEC; s_{ij}^T: isothermal elastic compliances; P: hydrostatic pressure ($\sigma_{ij} = -P\delta_{ij}$, where δ_{ij} is the Kronecker delta) or uniaxial pressure ($\sigma_{ij} = -Pt_it_j$, where t denotes the direction of uniaxial stress, $|t| = 1$); $g = s_{11}^T + 2s_{12}^T$.

$B_1 = -(1/g)\{1 + 2c_{11}^S[g + (1/f)(\partial f/\partial P)]\}$
$B_2 = -(1/g)\{1 + 2c_{44}^S[g + (1/f)(\partial f/\partial P)]\}$
$B_3 = -(3/g)\{1 + (2/3)(c_{11}^S + 4c_{44}^S + 2c_{12}^S)[g + (1/f)(\partial f/\partial P)]\}$
$B_4 = -(3/g)\{1 + (2/3)(c_{11}^S + c_{44}^S - c_{12}^S)[g + (1/f)(\partial f/\partial P)]\}$
$B_5 = -(2/g)\{1 + (c_{11}^S + 2c_{44}^S + c_{12}^S)[g + (1/f)(\partial f/\partial P)]\}$
$B_6 = -(1/g)\{1 + 2c_{44}^S[g + (1/f)(\partial f/\partial P)]\}$
$B_7 = -(2/g)\{1 + (c_{11}^S - c_{12}^S)[g + (1/f)(\partial f/\partial P)]\}$
$B_8 = -(1/s_{11}^T)\{1 + 2c_{11}^S[s_{11}^T + (1/f)(\partial f/\partial P)]\}$
$B_9 = -(1/s_{12}^T)\{1 + 2c_{44}^S[s_{12}^T + (1/f)(\partial f/\partial P)]\}$
$B_{10} = -(1/s_{12}^T)\{1 + 2c_{44}^S[s_{12}^T + (1/f)(\partial f/\partial P)]\}$
$B_{11} = -2c_{11}^S[1 + (1/s_{12}^T)(1/f)(\partial f/\partial P)]$
$B_{12} = -2c_{11}^S[1 + (1/s_{12}^T)(1/f)(\partial f/\partial P)]$
$B_{13} = -2(c_{44}^S/s_{12}^T)[s_{11}^T + (1/f)(\partial f/\partial P)]$
$B_{14} = -2(c_{44}^S/s_{12}^T)[s_{11}^T + (1/f)(\partial f/\partial P)]$
$B_{15} = -2(c_{44}^S/s_{12}^T)[s_{11}^T + (1/f)(\partial f/\partial P)]$
$B_{16} = -2c_{11}^S[1 + (1/s_{12}^T)(1/f)(\partial f/\partial P)]$
$B_{17} = -[4c_{44}^S/(s_{11}^T + s_{12}^T)][(1/2)(s_{11}^T + s_{12}^T + s_{44}^T/2) + (1/f)(\partial f/\partial P)]$

Table 3. *TOEC and pressure derivatives of SOEC of the aluminium alums of* K, NH_4, Cs *and* CH_3NH_3, *with limits of error in parentheses*

Units: TOEC (10^{11} dyn cm^{-2}); Pc_{ij} (10^{-5} bar^{-1}); Pc', Pc'' pressure derivatives of $c' = (c_{11} + c_{12} + 2c_{44})/2$, $c'' = (c_{11} - c_{12})/2$ respectively. All values for ca 22°C.

	$KAl(SO_4)_2 . 12H_2O$	$NH_4Al(SO_4)_2 . 12H_2O$	$CsAl(SO_4)_2 . 12H_2O$	$CH_3NH_3Al(SO_4)_2 . 12H_2O$
c_{111}	−22·2 (3%)	−7·5 (12%)	−21·2 (9%)	−23·7 (5%)
c_{112}	−7·1 (11%)	−1·1 (24%)	−11·1 (9%)	−11·9 (8%)
c_{113}	−8·6 (8%)	−2·0 (41%)	−12·6 (8%)	−11·5 (6%)
c_{123}	−13·4 (7%)	−1·9 (48%)	−9·0 (22%)	−10·4 (12%)
c_{144}	−2·3 (24%)	−2·9 (11%)	−2·7 (8%)	−1·1 (18%)
c_{155}	−8·02 (4%)	−5·62 (3%)	−5·90 (3%)	−4·30 (4%)
c_{166}	−7·44 (6%)	−4·94 (5%)	−5·36 (4%)	−2·93 (4%)
c_{456}	−2·0 (22%)	−0·64 (24%)	−1·6 (38%)	−0·61 (12%)
Pc_{11}	2·73 (4%)	0·4 (90%)	1·87 (5%)	2·01 (3%)
Pc_{12}	6·98 (4%)	1·7 (60%)	3·93 (6%)	3·50 (3%)
Pc_{44}	3·19 (5%)	2·07 (3%)	1·39 (2%)	0·394 (6%)
Pc'	3·94 (2%)	1·14 (4%)	2·36 (3%)	2·13 (4%)
Pc''	−0·038 (160%)	−0·78 (2%)	−0·155 (4%)	−0·075 (40%)

The relation follows from

$$c = \rho v^2 = \rho[2l(f_m - f_n)/(m - n)]^2.$$

This equation is used for the evaluation of any elastic constant c by resonance frequencies f_m of thick plane-parallel plates in non-piezoelectric crystals (Spangenberg & Haussühl, 1957).

Results

Table 3 presents the values for TOEC and pressure derivatives of SOEC together with limits of error as obtained from our least-squares computation.

All TOEC have negative signs, which is not a common feature of all other cubic crystals so far investigated, e.g. crystals of NaCl-type possess positive c_{123}, c_{144}, and c_{456}. Further, the differences $(c_{112} - c_{113})$ and $(c_{155} - c_{166})$, which vanish in symmetry group $4/m3$, exist in accordance with symmetry $m3$ of the alums. The structural relations between alums and CsCl-type crystals (Haussühl, 1961) suggest a similar nonlinear elastic behaviour in both crystal groups. For CsCl-type crystals, only calculated TOEC are available (Ghate, 1965). They all are negative too, and the constants with mixed indices c_{ijk} are much larger in magnitude than those of crystals of NaCl-type. The same holds for the alums. Another feature is the smallness of c_{111} compared with the values of other

Table 4. *Relations between SOEC and TOEC* (unit: 10^{11} dyn cm^{-2}) *for some selected crystals, at room temperature*

	c_{111}	c_{11}	c_{112}	c_{12}	c_{44}	c_{111}/c_{11}	c_{112}/c_{111}	Reference TOEC	SOEC
Cu	−127·1	16·61	−81·4	11·99	7·56	−7·65	0·640	a	a
Ag	−84·3	12·22	−52·9	9·07	4·54	−6·90	0·628	a	a
Au	−172·9	19·29	−92·2	16·38	4·15	−8·96	0·533	a	a
Si	−82·5	16·577	−45·1	6·392	7·962	−4·98	0·547	b	b
Ge	−71·0	12·853	−38·9	4·826	6·680	−5·52	0·548	b	b
LiF	−142	11·355	−26	4·76	6·35	−12·51	0·183	c	d
NaF	−148	9·70	−27	2·43	2·81	−15·26	0·182	e	d
KCl	−73	4·078	−2·4	0·69	0·633	−17·90	0·033	c	d
NaCl	−86·36	4·936	−4·96	1·29	1·265	−17·50	0·057	g	d
MgO	−489·5	28·917	−9·5	8·796	15·461	−16·93	0·019	h	i
CaF$_2$	−124·6	16·357	−40·0	4·401	3·392	−7·62	0·321	j	k
SrF$_2$	−82·1	12·461	−30·9	4·463	3·187	−6·59	0·377	l	l
BaF$_2$	−58·4	9·122	−29·9	4·148	2·551	−6·40	0·512	f	k
KAl(SO$_4$)$_2$.12H$_2$O	−22·2	2·465	−7·1	1·025	0·868	−9·01	0·320	m	m
NH$_4$Al(SO$_4$)$_2$.12H$_2$O	−7·5	2·520	−1·1	1·09	0·814	−2·98	0·147	m	m
CsAl(SO$_4$)$_2$.12H$_2$O	−21·2	3·115	−11·1	1·539	0·840	−6·81	0·524	m	m
CH$_3$NH$_3$Al(SO$_4$)$_2$.12H$_2$O	−23·7	2·971	−11·9	1·732	0·582	−7·98	0·502	m	m

References: (*a*) Hiki & Granato (1966). (*b*) McSkimin & Andreatch (1964). (*c*) Drabble & Strathen (1967). (*d*) Haussühl (1960). (*e*) Bensch (1972). (*f*) Gerlich (1968). (*g*) Swartz (1967). (*h*) Bogardus (1965). (*i*) Chung & Lawrence (1964). (*j*) Alterovitz & Gerlich (1969). (*k*) Haussühl (1963). (*l*) Alterovitz & Gerlich (1970). (*m*) Present work.

Table 5. *Pressure derivatives of SOEC* (unit: 10^{-5} bar^{-1}) *for some selected crystals at room temperature*

	Pc_{11}	Pc_{12}	Pc_{44}	Pc_{12}/Pc_{11}	Pc_{44}/Pc_{11}	Reference
CsCl	1·91	5·78	4·57	3·03	2·39	a
CsBr	2·19	6·44	5·06	2·94	2·31	a
CsI	2·74	7·68	6·11	2·80	2·23	a
LiF	0·88	0·57	0·22	0·65	0·25	b
NaF	1·19	0·84	0·073	0·71	0·06	b
KCl	3·17	2·29	−0·61	0·72	−0·19	c
NaCl	2·4	1·6	0·29	0·67	0·12	d
MgO	0·32	0·21	0·074	0·66	0·23	e
KAl(SO$_4$)$_2$.12H$_2$O	2·73	6·98	3·19	2·56	1·17	f
NH$_4$Al(SO$_4$)$_2$.12H$_2$O	0·4	1·7	2·07	4·25	5·18	f
CsAl(SO$_4$)$_2$.12H$_2$O	1·87	3·93	1·39	2·10	0·74	f
CH$_3$NH$_3$Al(SO$_4$)$_2$.12H$_2$O	2·01	3·50	0·394	1·74	0·20	f

References: (*a*) Chang & Barsch (1967). (*b*) Miller & Smith (1964). (*c*) Bartels & Schuele (1965). (*d*) Swartz (1967). (*e*) Anderson & Andreatch (1966). (*f*) Present work.

crystals. In Table 4 the constants c_{111}, c_{11}, c_{112}, and c_{12} are listed for different isotypic crystal groups. The ratios c_{111}/c_{11} and c_{112}/c_{111} vary only slightly within these isotypic groups. This should be considered as a general rule for stable crystals. A similar rule for thermoelastic constants $T_{ij} = \mathrm{d}\ln c_{ij}/\mathrm{d}T$ (T temperature) is known (Haussühl, 1963). The values c_{111}/c_{11} for α-KAl(SO$_4$)$_2$.12H$_2$O, β-CsAl(SO$_4$)$_2$.12H$_2$O and β-CH$_3$NH$_3$Al(SO$_4$)$_2$.12H$_2$O coincide with the corresponding quantities of crystals of Cu-type, diamond-type, and CaF$_2$-type. But for crystals of NaCl-type these ratios are about twice as large. The ratio c_{112}/c_{111} for all the crystals investigated so far does not differ strongly. The only exceptions are crystals of NaCl-type and α-NH$_4$Al(SO$_4$)$_2$.12H$_2$O with much smaller values. This unusual behaviour might be considered as a further manifestation of the intermediate position of NH$_4$ alums between normal α and β-alums. Thermal expansion and thermoelastic properties of α-NH$_4$ alums range between the values of stable α and β-alums (Haussühl, 1961).

The pressure derivatives Pc_{ij} show a much clearer difference between α and β-alums. Again the NH$_4$ alum possesses much lower values.

The pressure derivatives of alums agree well with the experimental data of caesium halides (Chang & Barsch, 1967), contrary to those of NaCl-type which exhibit completely different behaviour (Table 5).

We suggest that studies of TOEC may yield further valuable information about special contributions to anharmonic properties in crystals originating from asymmetric ions and molecules. Further investigations in this field are in progress.

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