THE COMPUTATION OF DYNAMICAL MODULI OF SOLIDS UNDER PRESSURE

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On the basis of the theory of finite strains, expressions are obtained in general form for the effective adiabatic second order elastic constants of crystals of any symmetry in terms of the isothermal elastic constants of second, third, and higher orders in the free energy decomposition. These expressions are used in the case of crystals of cubic symmetry under hydrostatic conditions to find the elastic wave velocities in mono- and polycrystals, and their pressure dependences. The polycrystal was considered as an isotropic body consisting of a large number of cubic monocrystals. The isotropic elastic constants were calculated from theoretical and experimental results for monocrystals in the Voigt-Reuss-Hill approximation. A method of applying this approximation to thermodynamic effective second order elastic constants is proposed. The results of a computation are compared with data of experiments to measure the sound velocity in polycrystalline NaCl and CsCl specimens under pressures to 100 kbar. The results of this comparison are discussed.

Investigations of the elastic properties of solids subjected to finite strains by dynamical methods result in the determination of the "effective" adiabatic elastic constants of a deformed mono- or polycrystal. At the same time, the isothermal elastic constants of second (VPU constants), third (TPU constants), and higher orders, determined for the initial, "natural," state of the monocrystal, are known from theoretical computations.

Let us find the connection between the effective adiabatic second order elastic constants of a solid and the isothermal elastic constants of a monocrystal in general form.

Let us distinguish three different states:

- 1) the original natural state characterized by the coordinates a_i and the density ρ_0 in which there are no stresses;
- 2) the "initial" strained equilibrium state characterized by the coordinates X_i and the density ρ^* ;
- 3) the state "at a given time" characterized by the coordinates x_i and the density ρ .

The components of the displacement vector from the initial state into the state at a given time are defined by the expressions

$$u_i = x_i - X_i$$

The connection between the mechanical and thermodynamical stresses is in conformity with Murnaghan [1]:

$$T_{ij} = \frac{1}{-J} \frac{\partial x_i}{\partial a_k} \frac{\partial x_j}{\partial a_m} t_{km}$$
(1)

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© 1974 Consultants Bureau, a division of Plenum Publishing Corporation, 227 West 17th Street, New York, N. Y. 10011. No part of this publication may be reproduced, stored in a retrieval system, or transmitted, in any form or by any means, electronic, mechanical, photocopying, microfilming, recording or otherwise, without written permission of the publisher. A copy of this article is available from the publisher for \$15.00. Here t_{km} are the thermodynamical stresses defined by the expressions

$$t_{km} = \rho_0 \, \frac{\partial U \left(\eta_{km}, S \right)}{\partial \eta_{km}} \,, \ t_{km} = \rho_0 \frac{\partial A \left(\eta_{km}, T \right)}{\partial \eta_{km}} \,, \ \rho = \frac{\rho_0}{J}$$

where $U(\eta, S)$ and $A(\eta, T)$ are the internal and Helmholtz free energies, respectively, η is the strain in Lagrange variables, S is the entropy, T is the temperature, J is the Jacobian of the transformation from the variables x_1, x_2, x_3 to the variables a_1, a_2, a_3 . Omitting second and higher order terms in the displacement gradients u_i [2], let us write the expression for the thermodynamical stresses as their series expansion near the initial state:

$$t_{km} = t_{km}^* + c_{kmpn}^* (\eta_{pn} - \eta_{pn}^*) + \ldots = t_{km}^* + c_{kmpn}^{*S} \frac{\partial X_I}{\partial a_p} \frac{\partial X_h}{\partial a_n} e_{fh}$$

$$e_{fh} = \frac{1}{2} \left(\frac{\partial u_f}{\partial X_h} + \frac{\partial u_h}{\partial X_f} \right)$$
(2)

According to (1) and (2), we obtain for the mechanical stress

$$T_{ij} = \frac{1}{J} \frac{\partial X_i}{\partial a_k} \frac{\partial X_j}{\partial a_m} t_{km} = T^*_{ij} \left(1 - \frac{\partial u_l}{\partial X_l} \right) + T_{il}^* \frac{\partial u_j}{\partial X_l} + T_{jl}^* \frac{\partial u_i}{\partial X_l} + C^{S}_{ijh} e_{lh}$$
(3)

where

$$T_{ij}^{*} = \frac{1}{J^{*}} \frac{\partial X_{i}}{\partial a_{\kappa}} \frac{\partial X_{j}}{\partial a_{m}} t_{\kappa m}^{*}$$
(4)

$$C_{ij/h}^{*S} = \frac{1}{J^{*}} \frac{\partial X_{i}}{\partial a_{m}} \frac{\partial X_{j}}{\partial a_{p}} \frac{\partial X_{f}}{\partial a_{q}} \frac{\partial X_{h}}{\partial a_{l}} c_{mpql}^{*S}$$
(5)

The asterisks denote quantities in the initial state.

From (3) we obtain the connection between the stresses and strains for the case of small adiabatic strains in the initial coordinates:

$$T_{ij}' = \left(\frac{\partial T_{ij}}{\partial e_{/h}}\right)_{\mathbf{S}, e=0} e_{/h} = C_{ij/h}^{*'\mathbf{S}} e_{/h}$$
(6)

where

$$C_{ij/h}^{i'S} = -T_{ij}^* \delta_{ih} + T_{if}^* \delta_{hj} + T_{jf}^* \delta_{ih} + C_{ijfh}^{S}$$
(7)

Henceforth, we shall consider only the case of homogeneous initial strain for which the quantities $C_{ijfh}^{*'S}$ are constants. Then, using (6), we obtain the wave equation

$$\rho^* \frac{\partial^2 u_i}{\partial \tau^2} = B_{ijfh}^* \frac{\partial^2 u_j}{\partial X_j \partial X_h}$$

$$B_{ijfh}^* = \delta_{ij} T_{jh}^* + \frac{1}{J^*} \frac{\partial X_i}{\partial a_m} \frac{\partial X_j}{\partial a_p} \frac{\partial X_j}{\partial a_q} \frac{\partial X_h}{\partial a_l} c_{mpql}^{*S}$$
(8)

where τ is the time.

Expanding the elastic constants c_{mpql}^{*S} in (7) and (8) into series near the natural state, we obtain

$$c_{ml\cdot ql}^{*\mathbf{S}} = c_{mpql}^{\mathbf{S}} + \left(rac{\partial c_{mpql}^{*\mathbf{S}}}{\partial \eta_{ns}}
ight)_{T} \eta_{ns} + \dots$$

where $(\partial c_{mpql}^{*S}/\partial \eta_{ns})_{T}$ are mixed third order elastic constants according to the Brugger definition [3]. Let us determine the effective isothermal second order elastic constants for which the following expressions are obtained:

$$C_{ijfh}^{*'T} = -T_{ij}^* \delta_{fh} + T_{if}^* \delta_{hj} + T_{jf}^* \delta_{ih} + \frac{1}{J^*} \frac{\partial X_i}{\partial a_m} \frac{\partial X_j}{\partial a_p} \frac{\partial X_j}{\partial a_q} \frac{\partial X_h}{\partial a_l} (c_{mpql}^T + C_{mpqlns}^T) \eta_{ns})$$
(9)

In order to calculate the dynamical elastic models of a solid it is necessary to relate the effective isotherm second order elastic constants to the effective adiabatic elastic constants of the same order, which are determined by (7).

Since the mechanical stress tensor can be represented by functions of the form

$$T_{ij} = T_{ij} \{\eta, e, T(\eta, e, S)\}$$

where η is the finite strain and e is an infinitesimal strain produced by a sound wave, then the effective adiabatic second order elastic constant is

$$C_{ijfh}^{**S} = \left(\frac{\partial T_{ij}}{\partial e_{fh}}\right)_{S,e=0} = \left(\frac{\partial T_{ij}}{\partial e_{fh}}\right)_{T,e=0} + \left(\frac{\partial T_{ij}}{\partial x_m}\right)_{n,e=0} \left(\frac{\partial t_{\star^m}}{\partial T}\right)_{n,e=0} \left(\frac{\partial T}{\partial e_{fh}}\right)_{S,e=0} = C_{ijfh}^{**T} + \frac{1}{J^*} \frac{\partial X_i}{\partial a_\kappa} \frac{\partial X_j}{\partial a_m} \frac{T \lambda_{\kappa m} \lambda_{fh}}{\rho_0 C_n^*} = C_{ijfh}^{**T} + \frac{1}{J^*} \frac{\partial X_i}{\partial a_\kappa} \frac{\partial X_j}{\partial a_m} \frac{T \alpha_{pn}^* c_{kmpn}^{*T} \alpha_{p'n'}^* c_{p'n'fh}^{*T}}{\rho_0 C_n^*} = \lambda_{ij} = \alpha_{vw}^* c_{vwij}^{*T}$$

Expanding λ_{ij} in a series near the natural state, we obtain

$$\lambda_{ij} = \lambda_{ij}(0) + \left(\frac{\partial \lambda_{ij}}{\partial \eta_{mn}}\right)_{T,\eta=0} \eta_{mn}$$

Here

$$\left(\frac{\partial \lambda_{ij}}{\partial \eta_{mn}}\right)_{T,\eta=0} = c_{ijvw}^T c_{zymn}^T \left(\frac{\partial s_{vwzy}}{\partial T}\right)_l + \alpha_{v,l} C_{ijvwmn}^T$$

Performing an analogous expansion, we have

$$C_{\eta}^{*}(\eta_{m_{n}}) = C_{\eta}(0) + \frac{T}{\rho_{0}} \left(\frac{\partial \lambda_{ij}}{\partial T}\right)_{\eta=0} \eta_{ij}$$

where s_{VWZY} are the isothermal second order elastic yield, $\alpha_{VW} = (\partial \eta_{VW} / \partial T)_t$ are the linear coefficients of thermal expansion, C_{η} the specific heat for constant volume.

Therefore

$$C_{ijfh}^{*'S} = -T_{ij}\delta_{fh} + T_{if}\delta_{hj} + T_{jf}\delta_{ih} +$$

$$+ \frac{1}{J^*} \frac{\partial X_i}{\partial a_m} \frac{\partial X_j}{\partial a_p} \frac{\partial X_f}{\partial a_a} \frac{\partial X_h}{\partial a_l} (c_{mpql}^T + C_{mpqlns}^T \eta_{us}) + \frac{1}{J^*} \frac{\partial X_i}{\partial a_k} \frac{\partial X_j}{\partial a_m} \frac{T \alpha_{pn}^* c_{kmpn}^{*T} \alpha_{p'n'}^* c_{p'n'fh}^{*T}}{\rho_0 C^* n}$$
(10)

This expression relates the effective adiabatic second order elastic constants to the isothermal second and third order elastic constants of a solid in the original state and is valid for an arbitrary kind of finite strain and crystals of all symmetry classes.

For the case of multilateral compression, high hydrostatic pressure, this expression simplifies.

In this case

$$T_{ij} = -\delta_{ij}P, \ \eta_{ij} = \eta\delta_{ij}, \ \frac{\partial X_i}{\partial a_m} = (1+2\eta)^{i/2}\delta_{im}$$
(11)

Using (5), (7), (11), we have for the effective second order elastic constant

$$C_{ijfh}^{*S} = P\left(\delta_{ij}\delta_{fh} - \delta_{ij}\delta_{hj} - \delta_{jj}\delta_{ih}\right) + (1 + 2\eta)^{1/2} c_{ijfh}^{*S}$$

$$c_{ijfh}^{*S} = c_{ijfh}^{S} + \left(\frac{\partial c_{ijfh}^{*S}}{\partial \eta_{vw}}\right)_{T} \eta_{vw} + \ldots = c_{ijfh}^{S} + \Gamma_{ij/hnn} \eta + \ldots$$

$$(12)$$

where Γ_{ijfhnn} are combinations of mixed third order elastic constants

$$P = -\eta \left(1 + 2\eta\right)^{-1/2} \left(\frac{1}{2} c_{iipp}^{T} + \eta \frac{C_{iippnn}^{T}}{6}\right)$$
(13)

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and finally

$$C_{ijfh}^{*'8} = P \left(\delta_{ij}\delta_{fh} - \delta_{if}\delta_{hj} - \delta_{jf}\delta_{ih}\right) + \left(1 + 2\eta\right)^{1/2} \left(c_{ijfh}^{T} + \eta C_{ijfhnn}^{T}\right) + \frac{T\lambda_{ij}\lambda_{fh}}{\rho_0 \left(1 + 2\eta\right)^{1/2} C_{\eta}^{*}}$$
(14)

which is valid for crystals of arbitrary symmetry.

The expressions (10) and (14) can naturally be used to compute the effective elastic constants of polycrystals under arbitrary deformations by starting from the isothermal second and third order elastic constants of a polycrystal in the original state.

However, in order to obtain the effective elastic constants of polycrystals by using the results of theoretical computations [4-8] for monocrystals, it is necessary to carry out a separate averaging of the isothermal second and third order elastic constants.

Averaging according to Voigt-Reuss-Hill (VRH approximation) [9] can be used for this purpose. Thus we can write for the second and third order elastic constants

$$c_{ijkl}^{\mathrm{H}} = \frac{1}{2} (C_{ijkl}^{\mathrm{V}} + C_{ijkl}^{\mathrm{R}})$$

$$c_{ijklmn}^{\mathrm{H}} = \frac{1}{2} (C_{ijklmn}^{\mathrm{V}} + C_{ijklmn}^{\mathrm{R}})$$

where C_{ijkl}^{H} and C_{ijklmn}^{H} are the isothermal second and third order elastic constants for a polycrystal averaged according to VRH, and c_{ijkl}^{V} , C_{ijklmn}^{V} and C_{ijkl}^{R} , C_{ijklmn}^{R} are the Voight [10] and Reuss [11] averages, respectively.

The following expressions [12]:

$$\begin{aligned} c_{ijkl}^{R} & {}_{klmn}^{K} = \frac{1}{2} \left(\delta_{im} \delta_{jn} + \delta_{in} \delta_{jm} \right) \\ c_{ijklmn}^{R} &= - c_{ijpo}^{R} c_{klrs}^{R} c_{mnuv}^{R} S_{pqrsuv}^{R} \end{aligned}$$

$$(15)$$

are valid in the case of Reuss averaging, where $s_{k/mn}^{R}$, s_{pqrsuv}^{R} are the second and third order yield moduli, respectively. The preceding expressions are also valid for monocrystals.

Thus, in the case of multilateral compression of polycrystals consisting of monocrystals of cubic symmetry, the adiabatic second order elastic constants obtained from data on the sound velocities under high pressure will equal for polycrystals

$$\langle C_{11}^{\prime S} \rangle = \langle B_{11}^{S} \rangle = \frac{v_{l}^{2} \rho_{0}}{(1+2\eta)^{s_{l}}} = K^{S} + \frac{4}{3} G = \langle c_{11}^{T} \rangle +$$

$$+ \eta \left(2 \langle c_{11}^{T} \rangle + 2 \langle c_{12}^{T} \rangle + \langle C_{111}^{T} \rangle + 2 \langle C_{112}^{T} \rangle \right) + \frac{T \langle \lambda_{11} \rangle^{2}}{\rho_{0} (1+2\eta)^{s_{l}} C^{*}_{\eta}}$$

$$\langle C_{44}^{\prime S} \rangle = \langle B_{44} \rangle = \frac{v_{l}^{2} \rho_{0}}{(1+2\eta)^{s_{l}}} = G = \langle c_{44}^{T} \rangle +$$

$$+ \eta \left(\langle c_{11}^{T} \rangle + 2 \langle c_{12}^{T} \rangle + \langle c_{44}^{T} \rangle + \langle C_{144}^{T} \rangle + 2 \langle C_{166}^{T} \rangle \right)$$
(16)

where K, G are the volume elasticity and shear moduli, v_l and v_t are the longitudinal and transverse sound velocities. The sign () shows that the quantities have been averaged and refer to polycrystals.

In conformity with (10) we have for the pressure

$$P = -\eta (1 + 2\eta)^{-\eta_2} [\langle c_{11}^T \rangle + 2 \langle c_{12}^T \rangle + \eta (\frac{1}{2} \langle C_{111}^T \rangle + 3 \langle C_{112}^T \rangle + \langle C_{123}^T \rangle)]$$

$$n = -P [\langle c_{11}^T \rangle + 2 \langle c_{12}^T \rangle$$

$$- (\frac{1}{2} \langle C_{111}^T \rangle + 3 \langle C_{112}^T \rangle + \langle C_{123}^T \rangle)] (\langle c_{11}^T \rangle + 2 \langle c_{12}^T \rangle)^{-3}$$
(18)

Let us show that we can arrive at the same averaging results by another means, which is substantially shorter in the case of crystals of cubic symmetry. This means is the direct averaging of the effective thermodynamic second order elastic constants for a monocrystal by the VRH approximation scheme.

We have from expressions of type (2)

$$\frac{\partial t_{km}}{\partial \beta_{pq}} \frac{\partial \beta_{pq}}{\partial t_{km}} = c^*_{kmpq} s_{pqk'm'} = \delta_{kk'} \delta_{mm'}$$







$$\beta_{pq} = \frac{\partial X_s}{\partial a_p} \frac{\partial X_t}{\partial a_q} e_{st}$$

$$e_{jhkm}^* = e_{jhk'm'}^* e_{kmpq} S_{k'm'pq}^*$$
(19)

Expanding this expression in a series near the natural state, we obtain

$$c_{jhkm} = c_{jhk'm'}c_{kmpq}s_{k'm'pq}$$
$$c_{jhkmvw} = -c_{jhk'm'}c_{kmpq}c_{vwst}S_{k'm'pqst}$$

which is analogous to (15). In the case of polycrystals we obtain similarly to the preceding

 $\langle c_{f \, b \, k \, m}^{*} \rangle = \langle c_{f \, h \, k' \, m'}^{*} \rangle \langle c_{k \, m \, p \, q}^{*} \rangle \langle s_{k' \, m' \, p \, q}^{*} \rangle$

Averaging by the VRH approximation scheme yields

$$\langle c^{*V}_{fhkm} \rangle = \frac{1}{2} (c^{*V}_{fhkm} + c^{*R}_{fhk'm'} c^{*R}_{kmpq} s^{*R}_{k'm'pq})$$

Expanding $\langle \mathbf{e}_{fhkm}^* \rangle$ near the natural state, we have

Considering the polycrystals $\langle c_{fhkm}\rangle$ and $\langle C_{fhkmvw}\rangle$ as means in the VRH approximation, we arrive at the expressions

$$\langle c_{fhkm}
angle = {}^{1}/{}_{2} \left(c_{fhkm}^{V} + c_{fhkm}^{R}
ight) \\ \langle C_{fhkmvw}
angle = {}^{1}/{}_{2} \left(c_{fhkmvw} - c_{fhk'mv}^{R} c_{kmpq}^{R} c_{vwst}^{R} S_{k'm'qst}
ight)$$

which are analogous to (15). Therefore, averaging the second and third order elastic constants ($\langle c_{fhkm} \rangle$, $\langle c_{fhkmvw} \rangle$) by the VRH approximation scheme in the calculation of the dynamic moduli of a polycrystal from the elastic constants of a monocrystal is equivalent to averaging the effective thermodynamic second order elastic constants $\langle c_{fhkm}^* \rangle$.

(20)

	Author	Curve number	4 11	Van der Waal's attractive force	Repulsive force				Agreement with experiment	
Material					first nei ghbors	not con- sidening sign of ion chain	consider- of ing sign of quantum sign of the sign of t	three particle interaction	v _l (P)	v _t (P)
NaCl	Nran'yan [4]	3	0.333		+	-		_	Poor	Poor
	Ghate [⁶]	4	0,288		+	+			Good	Good
	Lincoln et. al. [⁷]	5	0.317	+	÷	-	+		Good	Good
	Lincoln et. al. [⁷]	6	0.317	+	+	-	+	—	"Poor	Satis.
	Paul [8]	7	0.322	+		-		+	Satis.	Poor
CsCl	Nran' yan [⁵]	3	0.333		+-	-	_		Poor	Poor
	Ghate [6]	4	0.256	-	+	+	-	-	Good	Good
	Ghate [6]	5	0.333	_	+	+	_		Poor	Poor

Remark:

"+") taken into account

"-") not taken into account.

However, in the general case it is necessary to know the relation $\eta_{ij} = \eta_{ij} (T_{ij})$; i.e., it is necessary to average the second, third, etc. order elastic constants in order to calculate $\langle c_{fhkm}^* \rangle$. According to (13), for cubic monocrystals under hydrostatic compression conditions the relation $\eta_{ij} = \eta_{ij} (T_{ij})$ is

$$\eta = -P \left(c_{12}^T + 2c_{12}^T \right)^{-1} + P^2 \frac{\left[c_{11}^T + 2c_{12}^T - (\frac{1}{2}C_{111}^T + 3C_{112}^T + C_{123}^T) \right]}{(c_{11}^T + 2c_{12}^T)^3}$$

It is known [13] that in this case the combinations of elastic constants of a monocrystal $c_{11}^{T} + 2c_{12}^{T}$, $\frac{1}{2}C_{111}^{T} + 3C_{112}^{T} + C_{123}^{T}$ agree with their means in the VRH approximation in (18). This permits averaging the elastic constants of only second order $\langle c_{fhkm}^{*} \rangle$, which results in an abrupt cutoff in the calculations and affords the possibility of determining the polycrystalline dynamic moduli by means of the values of the effective thermodynamic elastic constants C_{11}^{*} , $C_{44}^{*'s}$, known from experiment, for a cubic monocrystal under hydrostatic compression conditions in the (001) and (110) directions.

Using the expressions obtained, we averaged the second and third order elastic constants determined experimentally for the monocrystals CsCl [14] and NaCl [15], and the pressure dependences of the propagation velocities for the longitudinal v_l and transverse v_t elastic waves were computed from (16), (18) in polycrystals of these materials.

These dependences (curves 2) are represented in Fig. 1 (for NaCl) and Fig. 2 (for CsCl) as compared with the analogous dependences for polycrystalline NaCl and CsCl specimens obtained experimentally by the authors ([15], curve 1).

The discrepancy for these dependences, the experimental for the polycrystals (curve 1) and that obtained by averaging from experiments on monocrystals (curve 2), does not exceed 3-4% for both NaCl and CsCl, which corresponds to the existing accuracy in determining the third order elastic constants and shows that the error of the averaging carried out is apparently less than the mentioned discrepancy.

Moreover, the pressure dependences of the propagation velocities of longitudinal and transverse waves for NaCl and CsCl polycrystals obtained by averaging the theoretically computed second and third order elastic constants are presented on these same figures.

Curve 3 in Fig. 1 is constructed from data in Nran'yan [4], curve 4 from Ghate [6], curves 5 and 6 from Lincoln et al. [7], and curve 7 from Paul [9].

Curve 3 in Fig. 2 is constructed from data in Nran'yan [4] and curve 5 from Ghate [6]. It should be noted that the temperature dependence of the elastic constants in [7] was computed by two methods. This

results in the appearance of two sets of third order elastic constants (for T = 298 °K) and, correspondingly, two dependences v(P) (curves 5 and 6).

The calculation of the third order elastic constants in the theoretical papers used [4-8, 17] was based on the central forces model with a Born-Meyer type potential in which the repulsion because of overlapping of the electron shells of the ions is

$$\varphi = B \exp(r/\varepsilon)$$

where B and ε are the repulsion parameters (ε is a "stiffness" coefficient).

The fundamental characteristics of the potentials [4-8] are presented in Table 1, where, taking account of the van der Waal's attraction, the second neighbors, and the three-particle interaction are noted. Estimates of the agreement with experiment are presented in the last columns of the table. The agreement is hence considered "good" if the computed theoretical dependence v(P) lies in the domain between curves 1 and 2 (Figs. 1 and 2) obtained from experiment, "satisfactory" if the deviation from the domain of curves 1-2 does not exceed the possible errors in the experiment, and "poor" if the dependence lies outside the mentioned region beyond the possible limits of the experimental errors. All the computations for v(P) were carried out to 50 kbar pressures for CsCl and 70 kbar for NaCl, since it is required to take account of the fourth and higher order elastic constants above these pressures for these materials [18].

The analysis of the computational results (Figs. 1 and 2 and the table) showed the essential influence of the nature of taking account of the instantaneous forces in the crystal on the third order elastic constants and, correspondingly, on the effective elastic constants and pressure dependences of the elastic wave propagation velocities.

The best results are obtained by taking account of the second neighbors and using the repulsion parameter and lattice constant determined at T = 298 °K and P = 0.

Taking account of three-particle interaction in the lattice potential plays a minor part.

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