

SVERIGES GEOLOGISKA UNDERSÖKNING

SER. C.

Avhandlingar och uppsatser.

N:o 460.

ÅRSBOK 38 (1944) N:o 2.

ON THE SUBSTITUTION
RELATIONS IN THE
AMPHIBOLE
GROUP

BY

NILS SUNDIUS



Pris kronor 0:50

STOCKHOLM 1944

KUNGL. BOKTRYCKERIET. P. A. NORSTEDT & SÖNER

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In his interesting paper on the crystal structure and chemical composition of the monoclinic amphiboles, B. E. Warren¹ has referred the chemical variations within this great mineral group to substitution processes within the space lattice deduced by him for tremolite. From rotation photographs of tremolite, actinolite, kupfferite, hornblende and grünerite he concludes that all these species may have essentially the same atomic structure and on the basis of analyses of the different species named and of some other hornblendes rich in R_2O_3 and R_2O he discusses the extent of the replacements occurring and the distribution of the different substituting elements among the cations in the tremolite. The replacements comprise a number of different elements and imply far-reaching exchanges of the positive ions in the tremolite. The processes involved include the following reactions: $OH \rightleftharpoons F$; $Ca \rightleftharpoons Mg, Fe, Mn$; $Ca \rightleftharpoons Na, K$; $Mg \rightleftharpoons Fe'', Al, Ti, Fe'''$ and Mn ; $Si \rightleftharpoons Al$. Special interest is attached to the statement that the variation in the amounts of the alkalies in hornblendes runs parallel with that of the substitution of Si by Al, this being due to the fact that the lower valence of the aluminium sets free negative valences in the oxygen ions that are neutralized by Na, K and Ca. In this case of substitution the atomic number for (Ca, Na, K) is raised from 2 to 3 at most, the latter being a boundary value, when the space rooms between the silicon tetrahedron chains are filled with positive ions.

Judging from Warren's paper all the atomic replacements discussed by him seem to be considered as collectively possible and as continuous. However, if this is right it would exclude the existence of distinct isomorphic series within the monoclinic amphibole group with definite boundaries against each other, and this does not agree with experience. As early as 1929 G. BESKOW made a statistical compilation of a great number of analyses of amphiboles and made the general statement that »the chemical areas of variation — in this group — are very restricted and separated from each other by great gaps of immiscibility».² For certain series of the group the same was shown somewhat later in greater detail by W. Kunitz³ and the present writer⁴. As a matter of interest it may be mentioned that from the chemical analyses available Kunitz deduced the same standard formula for R_2O_3 - and Na-, K-poor amphiboles as Warren found to be the right one in view of the structure.

¹ Z. Krist., Bd. 72, h. 5/6, p. 493, 1930.

² Transl. by the writer, S. G. U., Ser. C, No. 350, p. 230, 1929.

³ N. J., B. B., 60, 1939—30, p. 171.

⁴ Tscherm. Min. petrogr. Mitteil., 43, 1933, p. 422.

The question as to the solubility relations in the complicated amphibole group and the existence there of different isomorphic series being not only of great mineralogical interest but also of importance for the petrography and the microscopical identification, the matter will be taken up again here in a short discussion, principally on the basis of the three works referred to above, supplemented with some new facts.

The tremolite — actinolite series.

The Strahlstein-series forms a rather complete row of numbers extending from the nearly pure Mg-tremolite to the vicinity of the Fe-component. The corresponding substitution is a simple matter, as the size of the Mg- and Fe-atoms is similar. The substitution of the other positive ions, Ca and Si, by other elements seems to offer greater difficulties. This is especially the case with Ca. Its atomic size is rather great and it occupies a larger space in the lattice than Mg and is bounded to eight oxygene atoms against six for the latter. Mn is more intermediate in respect of size, and as a matter of fact we find that Ca is substituted by Mn to a considerable extent in many minerals. Warren attempts to escape the difficulties connected with the replacement of Ca in the tremolite by Mg or Fe by assuming that six of the oxygen atoms around Ca in the case of an exchange move closer and the other two are bound to two silicon each and have their valences satisfied. This is regarded as a reasonable and possible change.

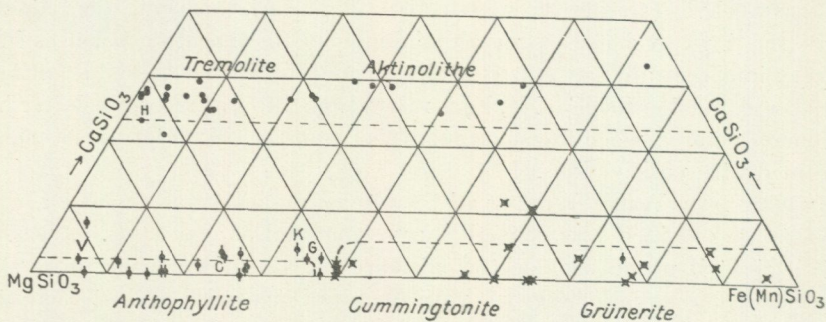


Fig. 1. Projection of analyses of cummingtonite-grünerite, anthophyllite and Strahlsteins in the $MgSiO_3$ - $Fe(Mn)SiO_3$ - $CaSiO_3$ -triangle (Mol. %). Points = Strahlsteins, points with vertical lines = anthophyllites, points with crosses = monoclinic Ca-poor amphiboles. Of the two points in the unmixing area between grünerites and Strahlsteins one is a mixture of two amphiboles, the other is contaminated by pyroxene.

However, the relations of the natural minerals do not agree with this assumption. In fact there is a distinct gap between the row of Strahlsteins and all Ca-poor amphiboles. For anthophyllites and Strahlsteins in a special case of paragenesis (Edwards, N.Y.) this gap was determined by the writer to extend from about 2 Mol. % $CaSiO_3$ in the anthophyllite to 23 Mol. % $CaSiO_3$ in the tremolite, and the extent of the unmixing area between grünerite-cummingtonite and Strahlsteins is not much smaller. As the theoretical content of CaO

in the Strahlsteins is about 28 Mol. % (water not considered), the boundary of the immiscibility does not lie far from the theoretical content. The extent to which Ca can be substituted by Mg and Fe is therefore rather restricted, which may depend on an inability of the lattice of tremolite-actinolite to sustain the deformation exercised by more than small amounts of replacement of this kind. The solubility relations are illustrated by the detailed diagram of fig. 1 by the writer and appears also in the more general diagram of Beskow in fig. 3—4.¹ In the rocks the immiscibility of both kinds of amphiboles is demonstrated by the paragenesis of members of both kinds in cases where equilibrium must be admitted. Both species of amphiboles, Ca-poor and Strahlsteins, also have been shown to form homoaxial intergrowths either in the form of lamellae or as mantles or terminal cappings. The question of the exchange of Si for Al in tremolite and actinolite will be discussed later.

Kupfferite and the Ca-poor amphiboles.

In an earlier paper dealing with the relations within the cummingtonite-grünerite series² the writer also made an investigation of the original kupfferite known from three localities in the Ural and Bajkal. Specimens of them were kindly placed at my disposal by Prof. Loevinsson-Lessing through the offices of Prof. H. Backlund. The kupfferite was first described as actinolite by v Kokscharoff; later on, because of an analysis by R. Herrmann in 1863, it has been considered as the type of a monoclinic Ca-poor amphibole approaching to the Mg-end of the cummingtonite-grünerite series. When investigating the three specimens I found that they were all similar and optically corresponded to an actinolite. Also the angle of the cleavage was in accord with this, the angle of actinolite being 55°36' against 54°20' for cummingtonite-grünerite. There is a distinct difference in this respect between the two kinds of amphibole. The actinolic character of the kupfferite was definitely proved by chemical determinations by A. Bygdén, repeated here for the sake of convenience.

	I	2
SiO ₂		57.46
TiO ₂	0.01	
Al ₂ O ₃	1.24	
Cr ₂ O ₃	0.34	1.21
FeO	3.67 ³	6.05
MnO	0.14	
NiO	0.13	0.65
MgO	22.27	30.88
CaO	12.69	2.94
Loss of ign.		0.81
		100.00

1. New determinations by Dr A. Bygdén.

2. Old analysis by R. Herrmann.

¹ The existence of an immiscibility area between the monoclinic Ca-poor amphiboles and the Strahlsteins has been established besides by the authors already named also by B. Askund (1925) and earlier by H. E. Johansson. Further references are given by G. Beskow.

² Am. J. Sc., XXI, 1931, 343.

³ Det. as Fe₂O₃.

According to the old analysis kuppferite is chemically related to the so-called amphibole-anthophyllite. From a research in 1931 of four specimens from Kongsberg received from Stockholm, Oslo, Trondheim and Vienna it appeared that they were all similar and were made up of a rhombic anthophyllite mixed with small amounts of a greenish or colourless actinolite. Also in this case a chemical analysis was made, showing a better accordance with the old ones. Thus kuppferite in the common sense of the name seems not to be represented in natural minerals. In the writer's paper of 1931, therefore, it was stated that in no case was a monoclinic Ca-poor amphibole richer in MgO than 60 mol. % known among the natural minerals. When the content of MgO is higher, an anthophyllite has been formed and representatives of this kind of amphiboles are well known. As a support of this view a case of paramorphism of both kinds of Ca-poor amphiboles was reported (Saude), where the two amphiboles evidently are in equilibrium with each other. When analysed separately, they were shown to differ by only about 2 or 3 mol. % MgSiO_3 , but the mixture is situated just beyond the boundary given above at 60 mol. % MgO. Hence the two kinds of Ca-poor amphiboles, the monoclinic and the rhombic, seem to form two rows in the continuation of each other, separated by a gap of immiscibility, though it is quite small (cp. the diagram of fig. 1).

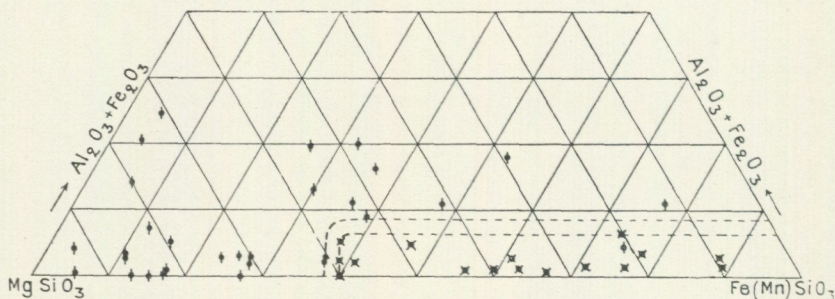


Fig. 2. Projection of analyses of Ca-poor amphiboles in the MgSiO_3 - Fe(Mn)SiO_3 - $\text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$ triangle (Mol. %). Points with vertical lines = anthophyllites and gedrites, points with crosses = monoclinic Ca-poor amphiboles.

Of course the relations described above hold good only when anthophyllites in the strict sense of the term are considered, i.e. if only members are included that are low in Al_2O_3 and Fe_2O_3 , the total amount of the two latter not exceeding 5–6 Mol. %. On the other hand, if gedrites with higher contents of these oxides are included, the relations change radically and the power of dissolving Fe⁺⁺ is greatly increased. Indeed, we know gedrites that are nearly as Fe⁺⁺-rich as grünerites. But the two series, the rhombic and the monoclinic, do not mix with each other (diagram of fig. 2).

The capacity of the rhombic modification of amphibole to take up sesquioxides and chiefly Al_2O_3 is a matter of great interest. At the same time the monoclinic Ca-poor members behave in quite a different manner and show a

much narrower limitation of their Al_2O_3 -absorbing capacity.¹ An excellent illustration hereof is afforded by a case of paragenesis of gedrite and cummingtonite in a specimen from Kalvola, Finland, described and discussed by P. Eskola.² Both amphiboles are mixed with each other and sometimes show intergrowths, the cummingtonite forming ends of the prisms of gedrite, the latter possibly in part being a little younger than the former. They were separated from each other and analysed. From the analyses I have calculated the number of metal atoms in the two amphiboles on the basis of the formula $(\text{OH})_2(\text{MgFe})_7\text{Si}_8\text{O}_{22}$ with the following result:

	Metal atoms and OH in the Cumming- tonite		Metal atoms and OH in the Gedrite		Ideal compos.
SiO_2	7.71	} 8.0	6.68	} 8.0	8.0
Al_2O_3	0.29		1.32		
Al_2O_3	0.01	} 7.09	0.64	} 7.16	7.0
TiO_2	0.04		0.06		
Fe_2O_3	0.48	} 7.09	0.41	} 7.16	7.0
$\text{Fe}(\text{Mn})\text{O}$	3.36		3.43		
MgO	3.20		2.62		
CaO	0.14	} 0.34	0.09	} 0.47	0
Na_2O	0.18		0.36		
K_2O	0.02		0.02		
H_2O	1.46		1.24		2

The most striking feature in the table is the considerable replacement of Si and (Fe, Mg) in the gedrite by Al, the corresponding substitution in the associated cummingtonite being small. In other respects the relations are similar, except for a certain increase in the Na-content of the gedrite, though it may be considered small when regarded as a consequence of the introduction of 1.32 atoms of Al in the Si-chains.

The facts related suggest that there is a causal connection between the capacity of the rhombic amphiboles to take up Fe" and the replacement of Si by Al. The corresponding space lattice must be regarded as rather different from the monoclinic one in cummingtonite and grünerite, which shows a greater rigidity. On the whole the pattern of the last-named amphiboles seems to be rather individual and displays more individual traces, both when compared with the rhombic species and with Strahlsteins. Thus the angle of the cleavage faces is $54^\circ 20'$ in the monoclinic Ca-poor members and $55^\circ 36'$ in the Strahlsteine and is given as $54^\circ 23' - 55^\circ 12'$ in the rhombic series. By K. Johansson³ it was stated that the parallelogram of a and c in the unit cell in the grünerite deviates from the rectangle of the anthophyllite by an angle of 4° as compared with $1^\circ 4'$ for actinolite. It is apparent also in the rotation photographs reproduced by Warren that the grünerite exhibits dissimilarities to all the other amphiboles photographed, whereas the latter are very similar to each other.

¹ In another connection B. Askund earlier has referred to the restricted capacity of cummingtonite to take up Al_2O_3 , S. G. U., Ser. C, No. 325, p. 31, 1925.

² Extrait des Comptes Rendus de la Soc. geol. de Finl., No. IX, 1936, p. 475.

³ Z. Krist., 73, h. 1, 1930, p. 31.

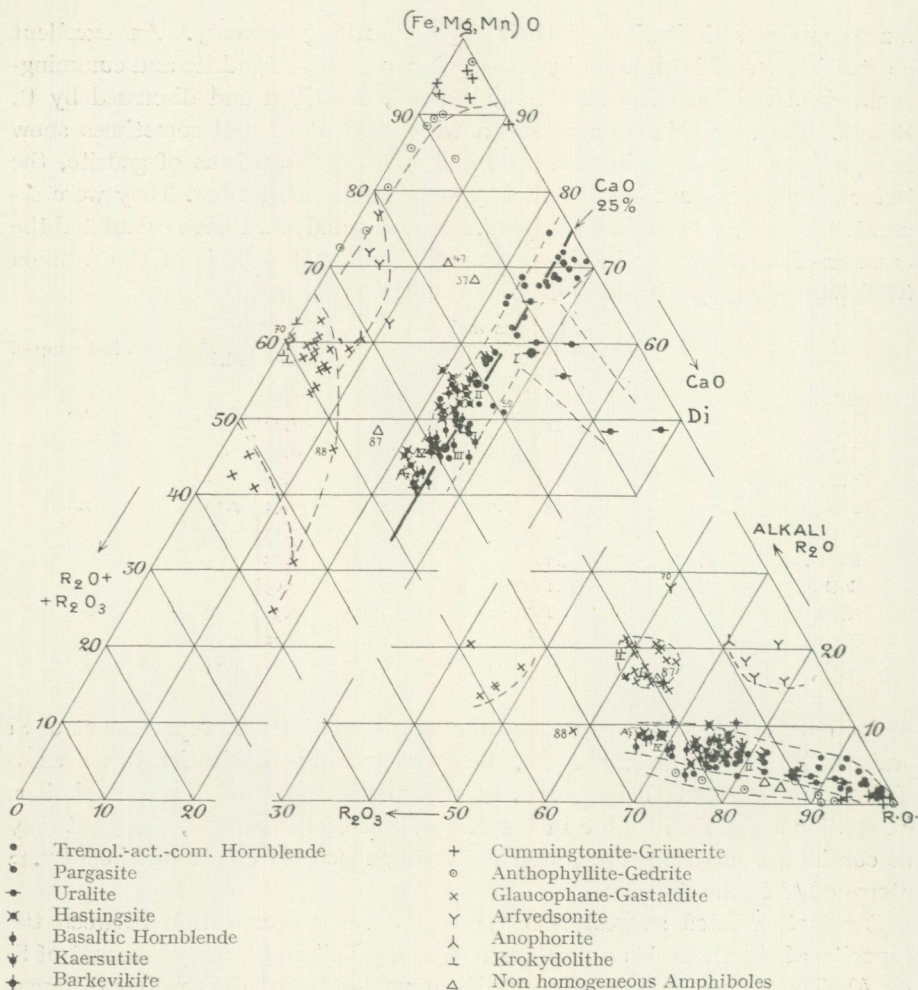


Fig. 3. The amphiboles in the two triangles $(\text{FeMnMg})\text{O} - \text{CaO} - (\text{R}_2\text{O} + \text{R}_2\text{O}_3)$ and $\text{RO} [= (\text{Fe}, \text{Mn}, \text{Mg}, \text{Ca})\text{O}] - \text{R}_2\text{O} (= \text{Alkalies}) - \text{R}_2\text{O}_3$. According to G. Beskow.

In ascribing to the monoclinic Ca-poor amphiboles full capacity of substitution of Mg by Fe, Warren bases his opinion on the photograph of an amphibole from Edwards, N.Y., designated as kupferite by Allen and Clement, who also give optical data of the mineral and reproduce an analysis of it. Specimens from the same locality have later been investigated by Kunitz and myself, and have in both cases been found to consist of a mixture of anthophyllite and tremolite. It was from this mixture that the unmixing area between the two kinds of amphibole was determined. Evidently the specimen used by Allen and Clement was identical with those studied by Kunitz and myself. According to their description it consists of two amphiboles, a tremolite and a prismatic and

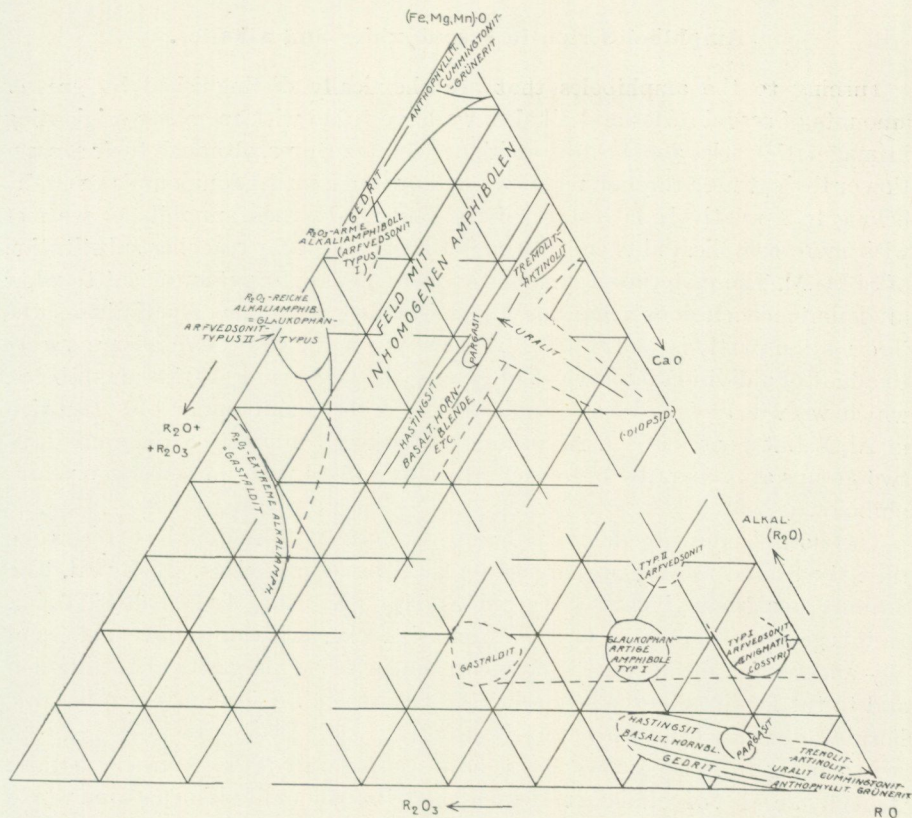


Fig. 4. The distribution of the different groups of Amphiboles in the triangles of fig. 3. According to G. Beskow.

fibrous one which »has an index of refraction $\gamma = 1.62$, and shows parallel extinction». This is in full accordance with that found for the anthophyllite (»Valleit») by Kunitz and myself. The mineral analysed by Allen and Clement and then used by Warren for the rotation-photograph is, therefore, not a monoclinic kupferite but an anthophyllite.

The three series discussed above, the anthophyllites-gedrites, the cummingtonites-grünerites and the Strahlsteins are all found in crystalline schists and may be designated as minerals formed at relatively low-temperature. Kunitz also referred glaucophane to the same category. Its formula is calculated by him as $H_2Na_2Mg_3Al_2Si_8O_{24}$ and according to the same author there is a close relationship between this amphibole and actinolite, but according to Beskow's general diagram (fig. 3—4) there is no transition between glaucophane and actinolite. The iron compound corresponding to glaucophane is riebeckite, $H_2Na_2Fe_3''Fe_2'''Si_8O_{24}$.

Amphiboles rich in sesquioxides and alkalis.

Turning to the amphiboles that are chemically distinguished by greater amounts of sesquioxides and alkalis we have to do with members originating in magmatic rocks and formed at higher temperatures, although their separation or their growth through reaction processes with earlier compounds generally belong to a relatively late stage of the cooling. In these amphiboles we met with more complicated chemical compositions and a far-reaching substitution of Si by Al. The material of systematic researches is not yet sufficient to allow of definite conclusions regarding the miscibility relations, especially as the information partly is contradictory. On the whole the respective minerals seem to admit of a division into two chief groups, the Ca-bearing Al-rich amphiboles, which we will designate as hornblendes, and the alkali-amphiboles also rich in Al_2O_3 but poor in CaO. According to Beskow's interesting diagram these two groups are separated from each other by a field with inhomogeneous amphiboles.

The hornblendes are distinguished by a normal Ca-content; at the same time the Si and (MgFe) are largely replaced by sesquioxides, chiefly Al. The formula has been given by Warren as $\text{H}_2(\text{Ca}, \text{Na}, \text{K})_{2-3}(\text{Mg}, \text{Fe}, \text{Al})_5(\text{SiAl})_8\text{O}_{24}$. In this group so important for the magmatic rocks, we seem to have a series of Mg- and Fe-rich members that may correspond to the pargasite at the Mg-end and to the hastingsite (hudsonite) at the Fe-rich end. Also the Ca-rich basaltic hornblendes may belong here.

According to Beskow the hornblendes are separated also from the Strahlsteins by an area of unmixing. The members that should fall in this area would be the so-called »common hornblendes», often reported from the rocks. However, safely proved instances of this kind are, according to Beskow, very rare. On the other hand, Kunitz considers a transition between the two groups probable.

The alkali-amphiboles are still less known from the point of view now considered, and will therefore, be touched upon but briefly. Apart from the Na-Al- and NaFe''' members glaucophane and ribeckite mentioned above a Na-Si-member corresponding to arfvedsonite should be quoted here. Kunitz gives for this species the general formula $\text{H}_2\text{Na}_2(\text{Mg}, \text{Fe}')_4\text{Si}_2\text{Si}_6\text{O}_{22}$ calculated from the analyses. As the analyses contain considerable amounts of Fe_2O_3 and Al_2O_3 Si and (MgFe) have probably to a certain degree been replaced by the two former. The formula is not in full accordance with the general amphibole-formula deduced by Warren and postulates a more noticeable departure of the atomic structure from that of tremolite.

Summary.

In the preceeding we have seen that the whole amphibole group comprises several isomorphic series which are separated by areas of unmixing. The series distinguished here are anthophyllite-gedrite, cummingtonite-grünerite, tremolite-actinolite, hornblendes (pargasite-hastingsite) and alkali-amphiboles. Considering the atomic structure the phenomenon of unmixing must be understood to be due to the existence of different patterns that cannot be transformed into each other without rebuilding the space lattice. In view of this it seems reasonable to suppose the existence of several subtypes of the amphibole structure deduced for tremolite, which may be fairly analogous but different enough to preclude the transition of one into another.

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