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The hierarchical internal structure of labradorite

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Supplementary 1: Derivation of the equations that give the An content or color of labradorescence depending on the thickness of the Ca-rich/Ca-poor lamellae.

Based on the work of Bolton et al. (1966) an equation was established by Miúra et al. (1975) that displays the origin of the respective color of labradorite. The wavelength λ of the reflected light is dependent on the thickness of a pair of lamellae d = (d_a + d_b), the refractive indice n, and the glancing angle θ. The total reflection amplitude V can then be calculated by:

$$V = r + \sum_{n=1}^{N}[1 - e^{i\beta_a(1+\gamma_a)}] * e^{i(n-1)}[\beta_a(1+\gamma_a) + \beta_b(1+\gamma_b)] \quad (4)$$

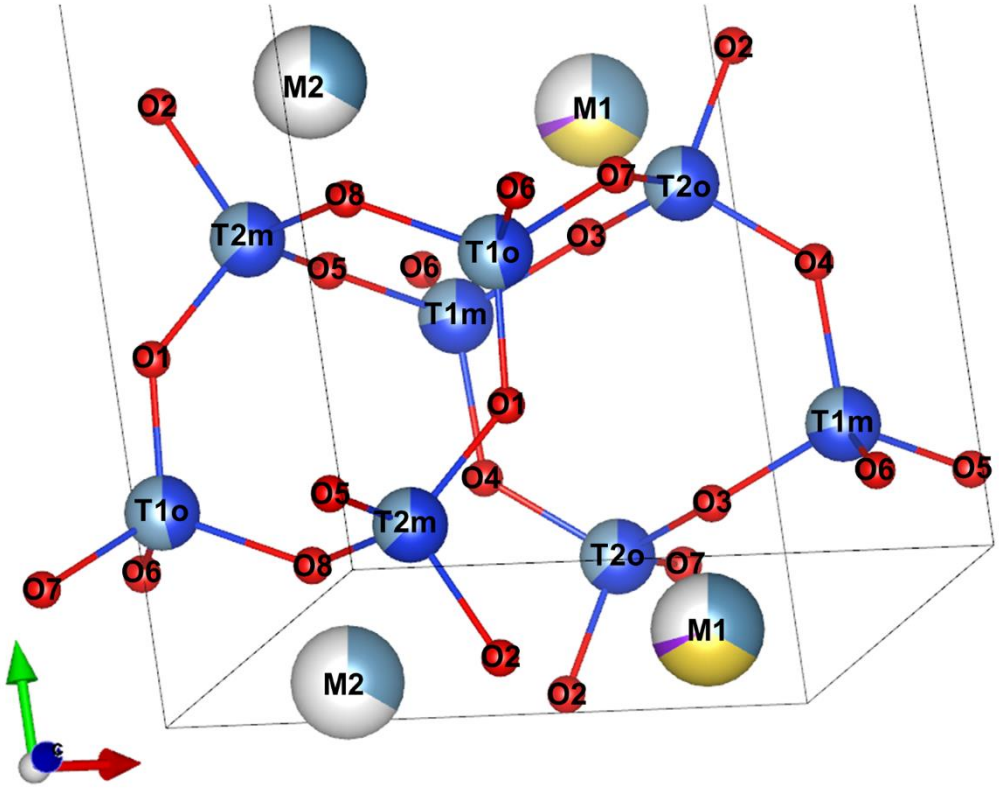
with the overall thickness of the crystal being N(d_a+d_b), the amplitude reflection coefficient r, and the Bragg equation for Ca- and Na- rich lamellae defining the term $\beta_m = 4\pi\lambda_m^{-1}n_m\sin\theta_m$, m = a, b and the distance $(1+\gamma_m)$ being a random variable of the symmetrical probability distribution of lamellae distances $f(\gamma)d\gamma$. If the lamellar thickness d is sufficiently small and N is sufficiently large, the observed average intensity $I = < |VV^*| > / r^2$ can be approximated by:

$$I \simeq \frac{e^{-2M}(1+e^{-2\alpha_a}) - 2e^{-(2M+\alpha_a)}\cos\beta_a}{1+e^{-2M} - 2e^{-M}\cos L} \quad (5)$$

Where $M = \alpha_a + \alpha_b$, $L = \beta_a + \beta_b$ and $-\alpha_m(\beta_m)$ is the cumulant of the characteristic function $f(\gamma_m)$ and is described by $< e^{i(\beta_m\gamma_m)} > = e^{-\alpha_m(\beta_m)}$.

A detailed derivation of the equations can be found in Bolton et al. (1966) and Miúra et al. (1975). Using Eq. (2), Miúra et al. (1975) calculated the reflected intensity I as a function of λ_m, d_m, n_m, θ_m and σ_m (standard deviations of mean thicknesses of alternate lamellae). Based on these parameters, regression equations for the wavelength λ and thus the An-content based on the lamellar thickness d were derived.

Supplementary 2: Part of the labradorite structure displaying the respective names of each atom.



Supplementary 3: T-O bond lengths of the single cell.

		Single cell		
		kinematical	dynamical	
T	O	Distance [Å]	Distance [Å]	Average [Å]
T2o	O2	1.66(2)	1.677(8)	1.652(7)
	O3	1.637(13)	1.633(5)	
	O4	1.610(17)	1.650(7)	
	O7	1.649(18)	1.646(7)	
T1o	O1	1.67(2)	1.653(9)	1.678(7)
	O6	1.700(17)	1.675(6)	
	O7	1.665(17)	1.686(7)	
	O8	1.668(14)	1.697(6)	
T1m	O3	1.620(13)	1.635(5)	1.646(8)
	O4	1.72(2)	1.671(10)	
	O5	1.650(19)	1.614(7)	

	O6	1.656(18)	1.664(7)	
T2m	O1	1.640(19)	1.649(7)	1.655(7)
	O2	1.68(2)	1.680(8)	
	O5	1.60(2)	1.641(7)	
	O8	1.664(13)	1.648(5)	
M1	O1	3.21(4)	3.651(18)	
	O2	2.34(2)	2.352(13)	
	O3	2.91(4)	3.570(18)	
	O4	3.12(4)	2.671(17)	
	O5	3.42(4)	2.730(16)	
	O6	2.48(4)	2.378(16)	
	O7	2.49(4)	2.422(16)	
	O8	2.40(4)	2.544(16)	
	M2	0.67(6)	0.78(2)	
M2	O1	3.60(4)	3.127(16)	
	O2	2.31(3)	2.310(10)	
	O3	3.58(5)	2.823(15)	
	O4	2.67(4)	3.190(17)	
	O5	2.79(4)	3.472(15)	
	O6	2.44(4)	2.470(13)	
	O7	2.42(4)	2.482(12)	
	O8	2.59(5)	2.405(15)	

Supplementary 4: Atom positions in the labradorite structure (xyz) of the single cell with their occupancy (occ) and their temperature factor (U(aniso); U11, U22, U33, U12, U13, U23) after kinematical refinement.

	x	y	z	occ	U(aniso) [Å]
M1Ca	-0.237(3)	0.492(4)	0.150(4)	0.32	-0.012(8) 0.29(5) 0.032(15) -0.032(17) 0.003(9) -0.14(2)
M1Na				0.33	
M1K				0.03	
M2Ca	-0.225(4)	0.528(3)	0.091(5)	0.32	0.001(14) 0.03(2) 0.025(19) 0.012(11) 0.022(12) 0.014(16)

T1oSi	0.0081(10)	0.1634(10)	0.2191(12)	0.46	-0.017(4) 0.030(9) -0.018(4) -0.004(4) -0.008(3) 0.001(4)
T1oAl				0.54	
T1mSi	0.0042(10)	0.8200(9)	0.2333(11)	0.69	-0.009(4) 0.015(8) -0.020(4) 0.007(4) -0.004(3) 0.000(4)
T1mAl				0.31	
T2oSi	0.1854(9)	0.6094(8)	0.3166(12)	0.64	-0.020(4) 0.009(8) -0.014(4) 0.000(4) -0.007(3) -0.005(4)
T2oAl				0.36	
T2mSi	0.1811(9)	0.3789(9)	0.3547(12)	0.62	-0.023(4) 0.039(10) -0.016(4) -0.001(4) -0.006(3) -0.004(4)
T2mAl				0.38	
O1	0.0142(19)	0.2915(14)	0.2762(19)	1	0.013(7) 0.004(13) -0.009(6) -0.004(6) -0.004(5) 0.001(6)
O2	0.0815(16)	0.4933(15)	0.2747(19)	1	-0.008(6) 0.033(15) -0.006(6) 0.001(6) -0.007(5) 0.007(7)
O3	0.3102(18)	0.6347(14)	0.5693(19)	1	0.007(7) 0.012(13) -0.013(7) 0.003(6) -0.012(5) -0.003(6)
O4	0.0170(15)	0.6867(15)	0.2181(18)	1	-0.012(6) 0.051(15) -0.016(6) 0.015(7) -0.015(5) -0.022(7)
O5	0.3148(17)	0.3506(17)	0.251(2)	1	-0.007(6) 0.066(17) 0.010(8) 0.005(7) 0.009(6) -0.014(8)
O6	-0.0012(18)	0.8685(15)	0.0202(17)	1	0.018(8) 0.051(15) -0.018(6) 0.009(8) 0.015(6) 0.014(7)
O7	0.3133(16)	0.6086(15)	0.1905(19)	1	-0.012(6) 0.043(15) -0.007(6) -0.025(7) -0.005(5) -0.009(7)
O8	0.3009(16)	0.3913(13)	0.6152(18)	1	-0.001(6) 0.001(12) -0.011(7) 0.000(6) -0.011(5) 0.004(6)

Supplementary 5: Atom positions in the labradorite structure (xyz) of the single cell with their occupancy (occ) and their temperature factor (U(aniso); U11, U22, U33, U12, U13, U23) after dynamical refinement.

	x	y	z	occ	U(aniso) [Å]
M1Ca	-0.2313(12)	0.4862(13)	0.1598(13)	0.32	0.027(3) 0.116(18) 0.051(6) 0.000(6) 0.016(4) -0.036(6)
M1Na				0.33	

M1K				0.03	
M2Ca	-0.2291(16)	0.5306(14)	0.0918(17)	0.32	0.013(4) 0.011(10) 0.013(5) 0.007(5) 0.005(3) 0.007(5)
T1oSi	0.0062(4)	0.1642(4)	0.2141(4)	0.46	0.0066(14) 0.016(4) 0.0087(16) -0.0054(18) 0.0022(12) -0.0010(18)
T1oAl				0.54	
T1mSi	0.0030(4)	0.8191(4)	0.2333(4)	0.69	0.0072(14) 0.008(4) 0.0092(15) 0.0084(17) 0.0029(11) 0.0011(17)
T1mAl				0.31	
T2oSi	0.1860(3)	0.6094(3)	0.3163(4)	0.64	0.0079(13) 0.000(4) 0.0089(15) 0.0048(16) 0.0030(11) 0.0015(16)
T2oAl				0.36	
T2mSi	0.1827(3)	0.3787(3)	0.3581(4)	0.62	0.0072(13) 0.008(4) 0.0086(15) -0.0008(17) 0.0021(11) 0.0027(17)
T2mAl				0.38	
O1	0.0147(6)	0.2908(6)	0.2767(7)	1	0.020(2) 0.045(8) 0.021(3) -0.010(3) 0.007(2) 0.003(3)
O2	0.0818(5)	0.4925(5)	0.2783(7)	1	0.015(2) 0.009(6) 0.019(2) -0.004(3) 0.0055(17) 0.001(3)
O3	0.3111(6)	0.6328(6)	0.5689(7)	1	0.019(2) 0.042(7) 0.014(2) 0.006(3) 0.0004(18) -0.003(3)
O4	0.0142(6)	0.6892(7)	0.2170(7)	1	0.021(2) 0.039(8) 0.022(3) 0.014(3) -0.0001(19) 0.000(3)
O5	0.3168(6)	0.3522(7)	0.2454(8)	1	0.024(2) 0.037(7) 0.041(3) 0.006(3) 0.020(2) -0.002(3)
O6	-0.0036(6)	0.8685(6)	0.0192(7)	1	0.028(2) 0.044(7) 0.013(2) -0.001(3) 0.0101(19) 0.004(3)
O7	0.3131(6)	0.6051(6)	0.1896(7)	1	0.022(2) 0.021(6) 0.023(2) -0.006(3) 0.0115(19) 0.002(3)
O8	0.3021(6)	0.3929(6)	0.6158(6)	1	0.020(2) 0.031(7) 0.013(2) -0.002(3) -0.0011(18) 0.006(3)

Supplementary 6: Position modulation waves of the labradorite structure.

	Position modulation		x	y	z

M1	Wave 1	cos	-0.005(2)	0.0266(17)	-0.0190(13)
		sin	-0.0342(18)	-0.0062(16)	0.0077(13)
	Wave 2	cos	0.001(2)	-0.0229(18)	0.0099(16)
		sin	0.010(2)	0.003(2)	0.0191(15)
M2	Wave 1	cos	-0.004(2)	0.009(2)	-0.0016(16)
		sin	0.014(4)	0.041(4)	-0.006(3)
	Wave2	cos	-0.020(4)	0.019(3)	-0.011(2)
		sin	-0.015(4)	0.012(3)	0.015(2)
T1o	Wave 1	cos	0.0013(14)	0.0071(12)	-0.0021(9)
		sin	0.0103(13)	0.0003(11)	0.0047(9)
	Wave 2	cos	-0.0009(14)	-0.0047(12)	0.0034(9)
		sin	0.0131(14)	0.0173(12)	0.0174(9)
T1m	Wave 1	cos	-0.0006(14)	-0.0017(12)	0.0015(9)
		sin	-0.0004(14)	0.0034(12)	-0.0030(9)
	Wave 2	cos	-0.0019(14)	0.0030(12)	0.0017(9)
		sin	-0.0104(14)	0.0085(12)	0.0116(9)
T2o	Wave 1	cos	-0.0020(13)	-0.0055(12)	0.0020(9)
		sin	0.0007(14)	0.0050(12)	0.0006(9)
	Wave 2	cos	0.0014(14)	-0.0078(12)	0.0034(9)
		sin	0.0067(14)	0.0159(12)	0.0195(9)
T2m	Wave 1	cos	-0.0003(14)	0.0033(12)	0.0007(10)
		sin	0.0037(15)	-0.0043(12)	0.0016(10)
	Wave 2	cos	-0.0068(15)	0.0038(13)	-0.0021(10)
		sin	0.0126(15)	0.0161(13)	0.0199(10)
O1	Wave 1	cos	-0.0018(19)	-0.0121(13)	-0.0034(13)

		sin	-0.0027(19)	-0.0075(15)	0.0028(12)
	Wave 2	cos	0.0300(19)	0.0043(16)	0.0072(13)
		sin	0.0065(19)	0.0061(15)	0.0225(12)
O2	Wave 1	cos	0.001(2)	0.0004(19)	-0.0005(15)
		sin	0.011(2)	0.0023(19)	0.0075(14)
	Wave 2	cos	-0.006(2)	-0.003(2)	-0.0003(15)
		sin	0.014(2)	0.0176(19)	0.0204(15)
O3	Wave 1	cos	-0.002(2)	0.0083(18)	-0.0001(15)
		sin	0.004(2)	-0.0033(17)	-0.0011(15)
	Wave 2	cos	-0.027(2)	0.0094(18)	-0.0109(15)
		sin	0.006(2)	0.0053(17)	0.0262(14)
O4	Wave 1	cos	-0.0060(18)	-0.0007(16)	0.0053(13)
		sin	0.0012(17)	0.0055(15)	-0.0022(13)
	Wave 2	cos	0.0006(17)	-0.0139(16)	0.0107(13)
		sin	-0.0092(17)	-0.0070(15)	0.0213(12)
O5	Wave 1	cos	0.001(2)	0.0131(19)	-0.0109(15)
		sin	0.004(2)	-0.001(2)	0.0015(15)
	Wave 2	cos	-0.013(2)	0.000(2)	-0.0022(17)
		sin	0.013(2)	0.017(2)	0.0195(16)
O6	Wave 1	cos	-0.002(2)	-0.0070(19)	-0.0035(16)
		sin	0.005(2)	-0.0041(19)	-0.0035(15)
	Wave 2	cos	0.007(2)	0.000(2)	0.0026(17)
		sin	0.026(2)	0.0158(19)	0.0259(16)
O7	Wave 1	cos	0.001(2)	-0.0117(18)	-0.0019(14)
		sin	0.000(2)	0.0073(18)	0.0026(14)

	Wave 2	cos	-0.020(2)	-0.0051(19)	0.0024(14)
		sin	-0.009(2)	0.0076(19)	0.0141(14)
O8	Wave 1	cos	0.003(2)	0.0026(19)	0.0028(15)
		sin	0.013(2)	-0.0030(19)	0.0057(15)
	Wave 2	cos	-0.002(2)	-0.003(2)	-0.0082(16)
		sin	0.019(2)	0.0231(19)	0.0229(15)

Supplementary 7: Occupancy modulation waves of the labradorite structure.

	Occupancy	Wave 1		Wave 2	
		ocos	osin	ocos	osin
M1Ca	0.32	-0.085(13)	0.114(11)	0.12(4)	0.29(2)
M1Na	0.33	0.085(13)	-0.114(11)	-0.12(4)	-0.29(2)
M1K	0.03	0.085(13)	-0.114(11)	-0.12(4)	-0.29(2)
M2Ca	0.32	-0.048(16)	-0.003(17)	0.19(2)	0.075(19)
T2oSi	0.64	0.096(13)	-0.106(12)	-0.06(4)	-0.31(2)
T2oAl	0.36	-0.096(13)	0.106(12)	0.06(4)	0.31(2)
T1oSi	0.46	0.081(13)	-0.117(11)	-0.14(4)	-0.29(2)
T1oAl	0.54	-0.08(13)	0.117(11)	0.14(4)	0.29(2)
T1mSi	0.69	0.100(3)	-0.101(12)	-0.03(4)	-0.32(2)
T1mAl	0.31	-0.100(3)	0.101(12)	0.03(4)	0.32(2)
T2mSi	0.62	0.086(13)	-0.114(11)	-0.11(4)	-0.30(2)
T2mAl	0.38	-0.086(13)	0.114(11)	0.11(4)	0.30(2)

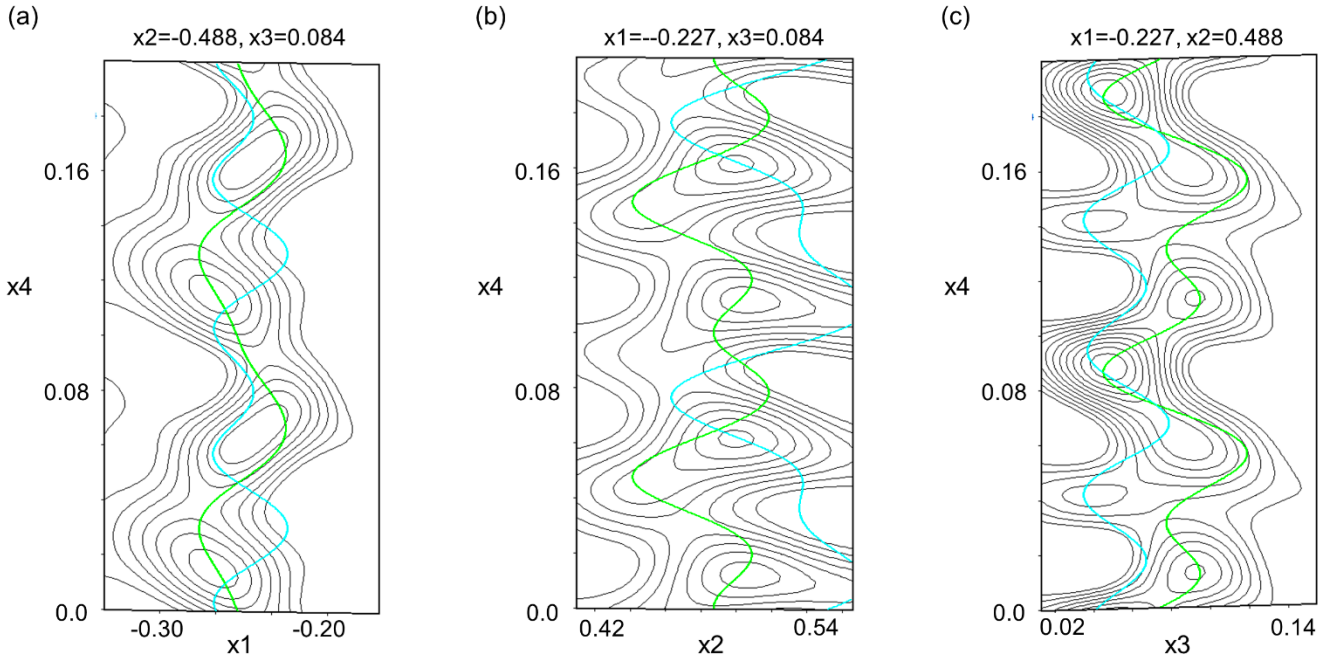
Supplementary 8: T-O bond lengths of the doubled cell.

	Double cell		
	kinematical		
T O	Average	Min. distance	Max. distance

		distance [Å]	[Å]	[Å]
T2o	O2	1.61(6)	1.49(6)	1.70(6)
	O3	1.59(4)	1.35(5)	1.83(5)
	O4	1.64(4)	1.49(5)	1.79(5)
	O7	1.60(6)	1.36(6)	1.82(6)
T1o	O1	1.62(5)	1.27(5)	1.99(5)
	O6	1.72(6)	1.62(6)	1.92(6)
	O7	1.73(5)	1.41(5)	2.03(5)
	O8	1.71(4)	1.66(5)	1.78(5)
T1m	O3	1.69(4)	1.49(4)	1.90(4)
	O4	1.73(5)	1.43(5)	2.05(5)
	O5	1.68(6)	1.56(6)	1.78(6)
	O6	1.68(6)	1.59(6)	1.77(6)
T2m	O1	1.67(5)	1.32(5)	1.96(5)
	O2	1.71(6)	1.59(6)	1.83(6)
	O5	1.63(6)	1.45(6)	1.73(6)
	O8	1.70(5)	1.54(5)	1.90(5)
M1	O1	3.16(6)	2.63(6)	3.55(6)
	O2	2.37(5)	1.99(6)	2.67(6)
	O3	2.78(7)	2.16(7)	3.48(7)
	O4	3.17(6)	2.73(6)	3.73(6)
	O5	3.57(7)	2.83(7)	4.34(7)
	O6	2.42(6)	2.17(6)	2.99(6)
	O7	2.61(6)	2.31(6)	2.81(6)
	O8	2.37(7)	1.99(7)	2.63(7)
	M2	1.02(9)	0.37(11)	1.58(11)
M2	O1	3.62(9)	2.71(10)	4.39(10)
	O2	2.35(7)	2.07(8)	2.67(8)
	O3	3.52(10)	2.94(10)	3.95(10)
	O4	2.69(8)	2.19(10)	3.42(10)
	O5	2.83(9)	2.20(10)	3.54(10)
	O6	2.43(3)	1.96(9)	2.91(9)

O7	2.52(8)	2.28(9)	2.71(9)
O8	2.48(9)	2.40(10)	2.67(10)

Supplementary 9: De Wolff's sections of the two M positions of the modulated structure. M1 is displayed in green, M2 in cyan. (a)-(c) show x4 vs. x1, x2 and x3, respectively.



Supplementary 10: Coordinates (a) δh, (b) δk, (c) δl and (d) period of the modulation vector *t* plotted against the An content. The values of the labradorite studied here are displayed as a red dot and agree with the ranges given for an An content of 53 mol%. The values predominantly follow the e_α-line. (e_α- and e_β-line after Jin et al. (2020)).