(ii) Hexagonal $BaTiO_3$

1a The hexagonal $BaTiO_3$ can be produced by quenching but quite stable below about 1000°C. [a]) In 1985, Sawaguchi, Akishige and Kobayashi found the ferroelectric activity below –199 °C. [b]) [a])60Gla [b])85Saw1

b

phase	III [a])	II [a])	I [a])
state	F [a])	P	P
crystal system	monoclinic [d])	orthorhombic [c])	hexagonal [b])
space group	$P2_1 - C_2^2$ [d])	$C2221 - D_2^5$ [e])	$P6_3/mmc - D_{6h}^4$ [b])
Θ [°C]	–199	–51	

[a])85Saw1 [b])48Eva [c])88Aki1 [d])90Saw [e])88Yam1

P_s || [001] of phase I.
Hexagonal form (phase I): $\rho = 5.865 \cdot 10^3$ kg m^{-3} (calculated from lattice constants); see 94Aki1
Transparent and pale yellow: see 86Aki

2a Crystal growth:
1) flux method (flux K_2CO_3 and Na_2CO_3 [a]), or K_2CO_3 and KF [b])); see [a]) 48Bur [b]) 80Shi
2) cooling method; see 85Saw1
3) floating zone method; see 87Yam
Thin film growth: evaporation on cleavage surfaces of NaCl; see 84Shi

b Crystal forms: Fig. 1A-10-240, Fig. 1A-10-241.

3a Unit cell parameters:
phase I: $a_h = 5.723$ Å $\approx \sqrt{2}\ a_c$, $c_h = 13.965$ Å $\approx 2\ \sqrt{3}\ a_c$ at 20 °C; 86Aki, 48Eva
(a_c: lattice parameter of cubic perovskite).
Phase II: $a = 5.723$ Å, $b = 9.898$ Å, $c = 13.946$ Å at –123 °C; 88Aki1
($a \approx a_h$, $b \approx \sqrt{3}\ a_h$, $c \approx c_h$).

b Crystal structure:
phase I: $Z = 6$ [d]), [d]) 48Bur
Table 1A-10-031, Table 1A-10-032; Fig. 1A-10-242, Fig. 1A-10-243;
see also 81Fis

4a Thermal expansion:
Fig. 1A-10-244, Fig. 1A-10 –245; Table 1A-10-033.
Lattice distortion due to p: Fig. 1A-10-246;
bulk modulus is $1.10(5) \cdot 10^{11}$ N m^{-2}; see 88Aki2

5a Dielectric constant vs. T : Figs. 1A-10-247...1A-10-251.
Dielectric dispersion: Fig. 1A-10-252, Fig. 1A-10-253.
Effect of hydrostatic pressures p on κ: Fig. 1A-10-254, Fig. 1A-10-255.
Phase diagram in regard to p: Fig. 1A-10-256, Fig. 1A-10-257.
Effect of unaxial pressure on κ near Θ_{II-I}: Fig. 1A-10-258, Fig. 1A-10-259.
Effect of E_{bias} on κ near Θ_{III-II}: Fig. 1A-10-260.

c Spontaneous polarization: Fig. 1A-10-261.

6a	Heat capacity: Fig. 1A-10-262; ΔQ_m = 220 J mol^{-1}, ΔS_m = 1.2 J mol^{-1}K^{-1} at $\Theta_{II–I}$; see	88Aki3
8a	Elastic compliances: Fig. 1A-10-263.	
9a	Refractive indices: Table 1A-10-034; Fig. 1A-10-264. Birefringence: Table 1A-10-034; Fig. 1A-10-265, Fig. 1A-10-266. Absorption edge: Fig. 1A-10-267; Temperature dependence of the absorption edge; see	95Aki1
b	Quadratic electrooptic constant: $M_{33} - M_{13}$ = 0.12(2) m^{4}C^{-2}, $M_{33} \approx$ 0.1 m^{4}C^{-2}.	87Aki
e	Optical SHG appears below $\Theta_{II–I}$: Fig. 1A-10-268.	
10a	Raman scattering: Figs. 1A-10-269...1A-10-273. Hydrostatic pressure effect on the Raman spectra and mode-Grüneisen parameters: see Hyper-Raman scattering: Figs. 1A-10-274...1A-10-277.	91Aki
b	Brillouin scattering: Fig. 1A-10-278.	
11	Electrical resistivity in reduced crystal: Fig. 1A-10-279. Luminescence of Eu^{3+}; see	94Ohi
13b	ESR: Fig. 1A-10-280; Table 1A-10-035.	
14a	Bragg reflections due to structural modulation: Fig. 1A-10-281, Fig. 1A-10-282.	
15	Twin structure in phase II: see	89Aki, 89Yam
	Twin structure in phase III: see	90Saw

Table 1A-10-031. $BaTiO_3$ (hexagonal). Fractional coordinates of atoms in the unit cell of phase I [94Aki1]. B_{eq}: equivalent isotropic displacement parameter.

Atom	*x*	*y*	*z*	B_{eq} [Å^2]
Ba(1)	0	0	1/4	0.43
Ba(2)	1/3	2/3	0.09671(5)	0.52
Ti(1)	0	0	0	0.67
Ti(2)	1/3	2/3	0.84633(14)	0.62
O(1)	0.5185(6)	0.0370	1/4	0.65
O(2)	0.8349(6)	0.6698	0.0802(2)	0.66

Table 1A-10-032. $BaTiO_3$ (hexagonal). Interatomic bond lengths and bond angles in phase I [94Aki1].

	Bond length [Å]		Bond angle [°]
$Ti(1)O_6$ octahedron			
O(2)-O(2)	3.965(4)×3	O(2)-Ti(1)-O(2)	180.00(1)
O(2)-O(2)	2.8334(3)×6	O(2)-Ti(1)-O(2)	91.2(1)
O(2)-O(2)	2.774(7)×6	O(2)-Ti(1)-O(2)	88.8(2)
Ti(1)-O(2)	1.983(2)×6		
Ti(1)-Ti(2)	3.940(1)		
$Ti(2)O_6$ octahedron			
O(1)-O(2)	3.932(2)×3	O(1)-Ti(2)-O(2)	169.1(1)
O(1)-O(1)	2.5434(3)×3	O(1)-Ti(2)-O(1)	79.37(6)
O(2)-O(2)	2.8904(3)×3	O(2)-Ti(2)-O(2)	95.1(1)
O(1)-O(2)	2.848(4)×6	O(1)-Ti(2)-O(2)	92.3(1)
Ti(2)-O(1)	1.991(1)×3		
Ti(2)-O(2)	1.959(2)×3		
Ti(2)-Ti(2)	2.690(4)		
Ba-O cuboctahedra			
Ba(1)-O(1)	2.868(4)×6	Ba(2)-O(1)	2.820(6)×3
Ba(1)-O(2)	2.881(3)×6	Ba(2)-O(2)	2.871(4)×6
		Ba(2)-O(2)	2.981(3)×3

Table 1A-10-033. $BaTiO_3$ (hexagonal). Linear thermal expansion coefficients α_i and volume expansion coefficient β_V for phase I and phase II [88Aki1].

Orthorhombic (phase II)				Hexagonal (phase I)		
α_a $[\cdot 10^{-6}K^{-1}]$	α_b	α_c	β_V	α_a $[\cdot 10^{-6}K^{-1}]$	α_c	β_V
−11.0(5)	14.0(10)	9.8(2)	12.0(10)	10.6(3)	9.8(2)	31.0(5)

Table 1A-10-034. $BaTiO_3$ (hexagonal). Refractive indices at λ = 0.633 μm [86Aki].

	Perovskite $BaTiO_3$ [a])	Hexagonal $BaTiO_3$
n_a	2.41	2.31(2)
n_c	2.36	2.39(2)
Δn	−0.053	0.08(1)

[a]) [68Wem] at RT.

Table 1A-10-035. $BaTiO_3 : Fe^{3+}$ (hexagonal). Spin Hamiltonian parameters of Fe^{3+} substituting for Ti^{4+} ion and Eulerian angles of the principal axes of the crystalline field tensor referenced to the crystallographic axes in the room temperature hexagonal phase [90Shi].

T	FS [$\cdot 10^{-2}$ m^{-1}]					Eulerian angles [°]
[K]	D	E	b_{40}	b_{42}	b_{44}	(α, β, γ)
283	–580	3	45	15	53	(90, 3, –90)
173	–610	46	47	–85	–120	(–92, 5, 0)
77	–630	120	42	200	–120	(–90, 10, 0)
63	–630	120	52	–27	12	(–129, 11, 63)
5	–620	89	41	120	41	(–150, 6, 90)

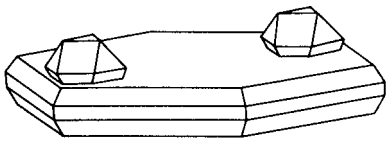

Fig. 1A-10-240. $BaTiO_3$ (hexagonal). Crystal form of hexagonal $BaTiO_3$ and coalescence of octahedral faces of small cubic crystals on hexagonal basal face [48Bur].

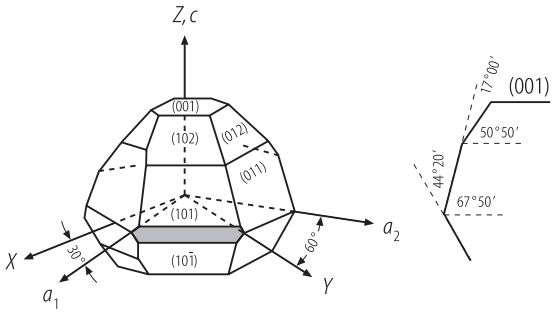

Fig. 1A-10-241. $BaTiO_3$ (hexagonal). Morphology of the single crystal grown by a flux method [80Shi]. Shaded area shows a rough surface.

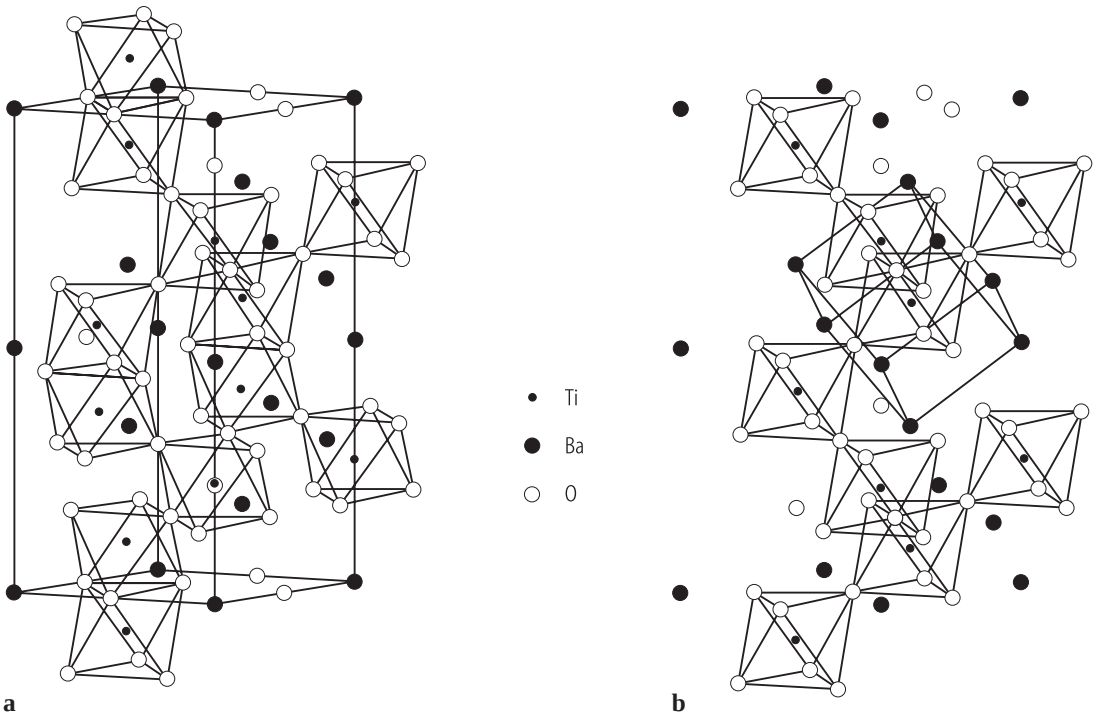

Fig. 1A-10-242. $BaTiO_3$ (hexagonal). Crystal structures of two polymorphic forms of $BaTiO_3$ [48Bur]; **(a)**: hexagonal, **(b)**: cubic. The hexagonal *c*-axis corresponds to the [111] direction of the cubic structure.

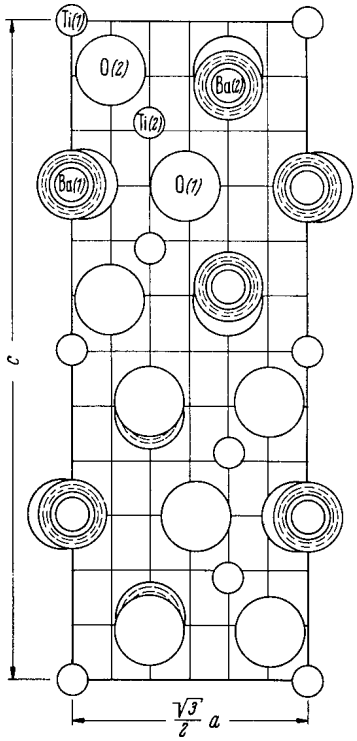

Fig. 1A-10-243. $BaTiO_3$ (hexagonal). Projection of hexagonal structure on (010) [48Bur].

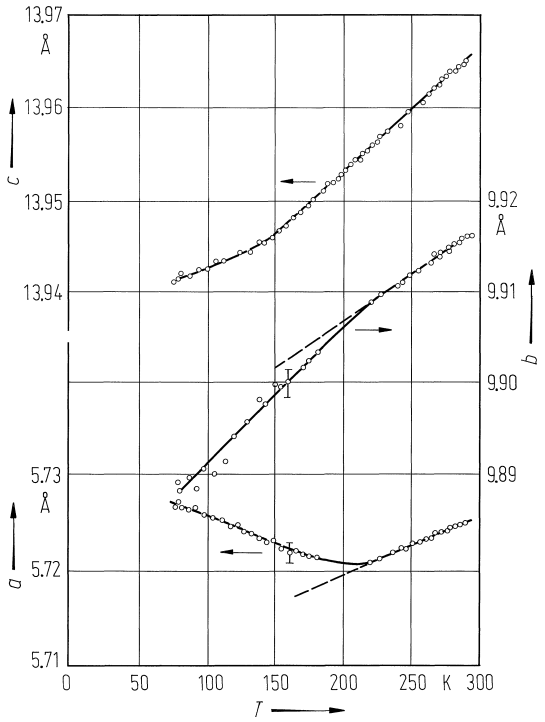

Fig. 1A-10-244. $BaTiO_3$ (hexagonal). *a*, *b*, *c* vs. *T* [88Aki1]. *a*, *b*, *c*: unit cell parameters of orthorhombic phase (phase II).

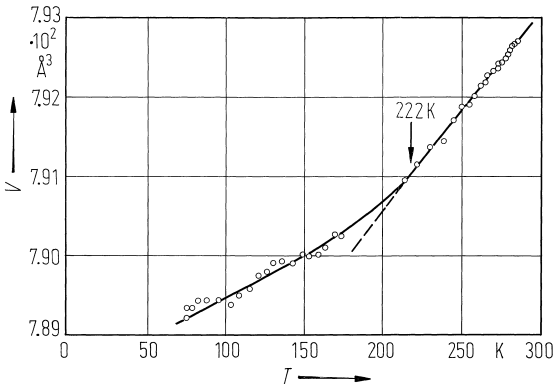

Fig. 1A-10-245. $BaTiO_3$ (hexagonal). *V* vs. *T* [88Aki1].
V: unit cell volume.

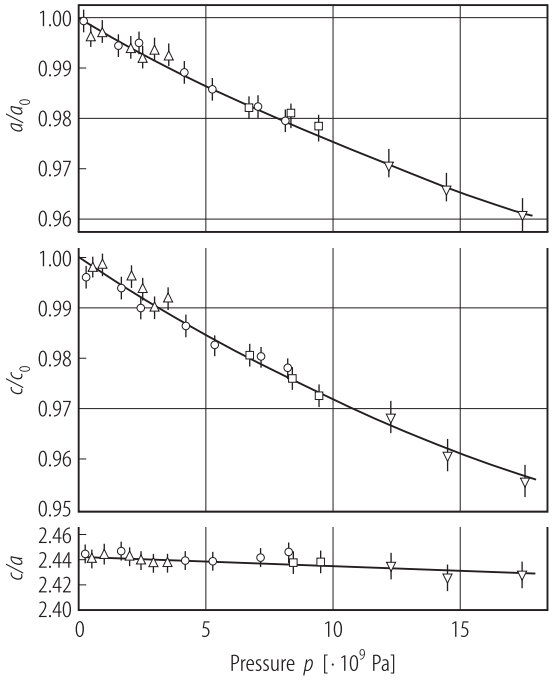

Fig. 1A-10-246. $BaTiO_3$ (hexagonal). a/a_0, c/c_0 and c/a vs. p [88Aki2]. a_0, c_0: unit cell parameters at 1 atm. p: hydrostatic pressure.

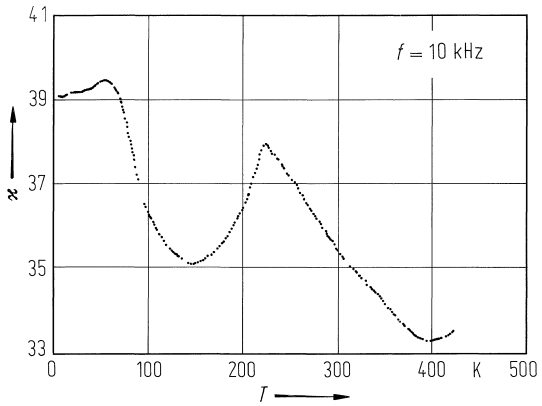

Fig. 1A-10-247. $BaTiO_3$ (hexagonal, ceramics). κ vs. T [85Saw2]. f = 10 kHz.

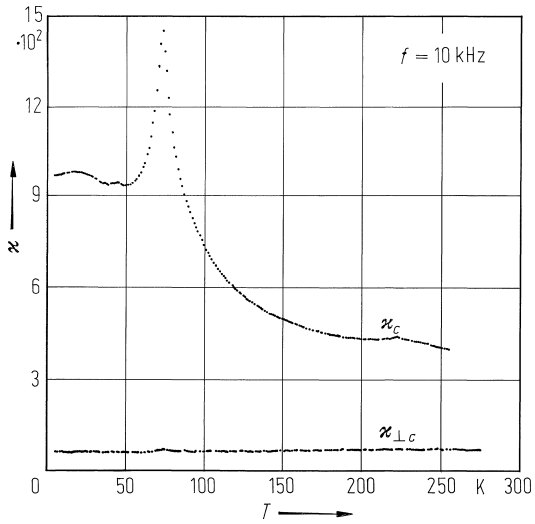

Fig. 1A-10-248. $BaTiO_3$ (hexagonal). κ_c, $\kappa_{\perp c}$ vs. T [85Saw1]. $\kappa_{\perp c}$: dielectric constant perpendicular to the c-axis. f = 10 kHz.

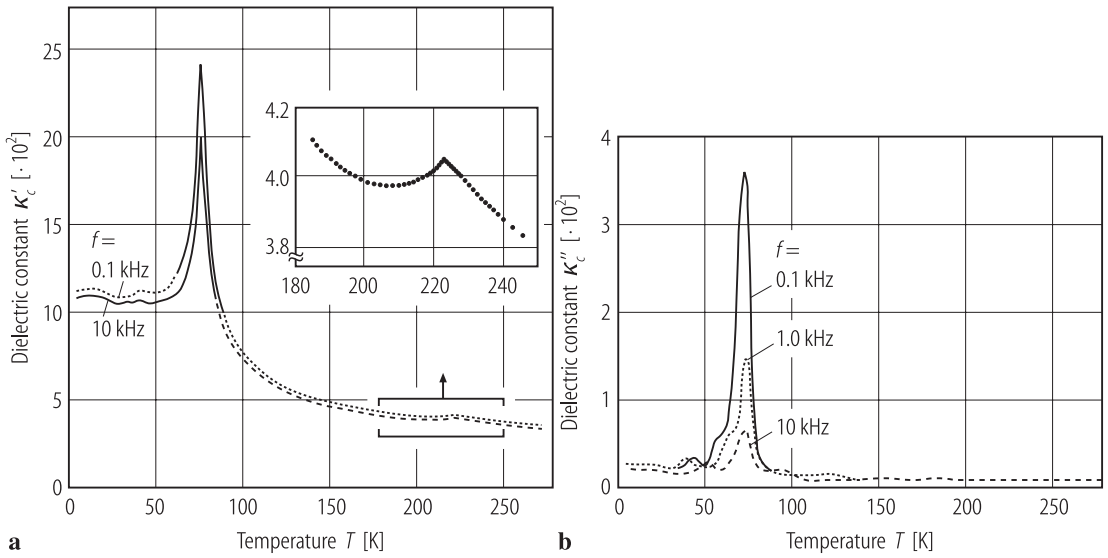

Fig. 1A-10-249. $BaTiO_3$ (hexagonal). κ_c' (**a**), κ_c'' (**b**) vs. T [89Aki]. Parameter: f. On heating.

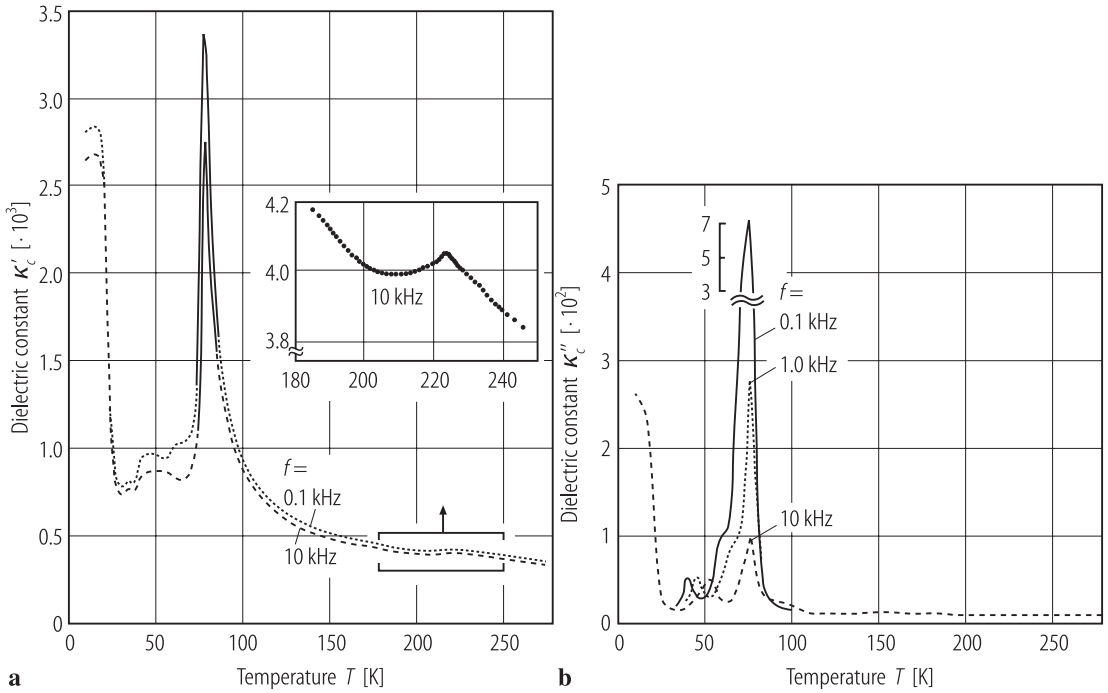

Fig. 1A-10-250. $BaTiO_3$ (hexagonal). κ_c' (**a**), κ_c'' (**b**) vs. T [89Aki]. Parameter: f. On heating after an electric field cooling from RT to 4.2 K under $E_{bias} = 1.5\ MV\,m^{-1}$.

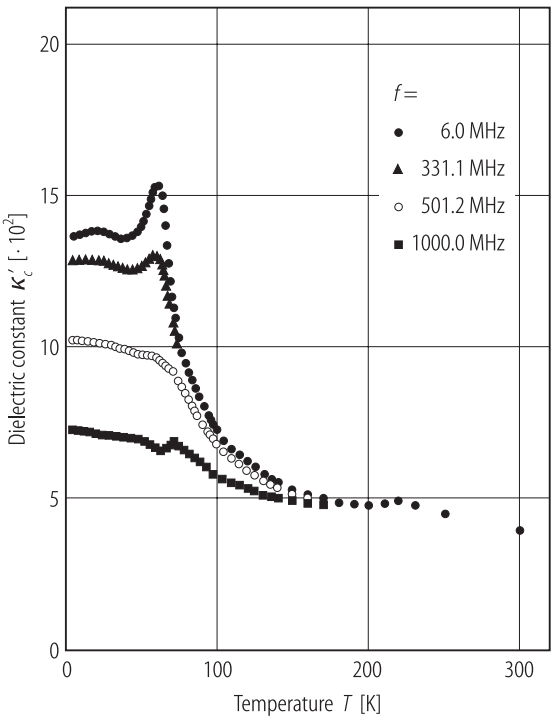

Fig. 1A-10-251. $BaTiO_3$ (hexagonal). κ_c' vs. T [88Deg]. Parameter: f.

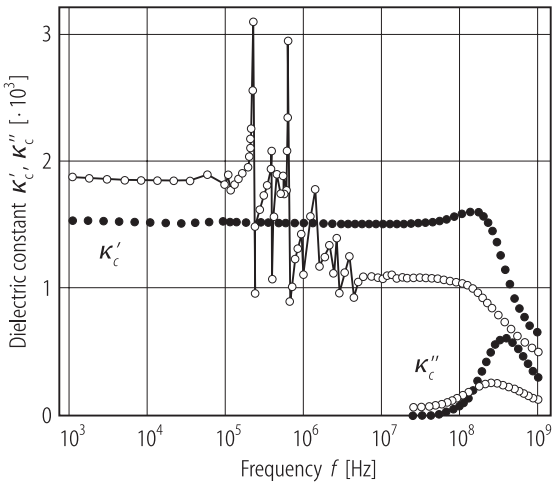

Fig. 1A-10-252. $BaTiO_3$ (hexagonal). κ_c', κ_c'' vs. f [88Deg]. T = 63 K. Full circle: after zero field cooling. Open circle: after field cooling of E_{bias} = 1 M V m^{-1} from 210 K to 74 K.

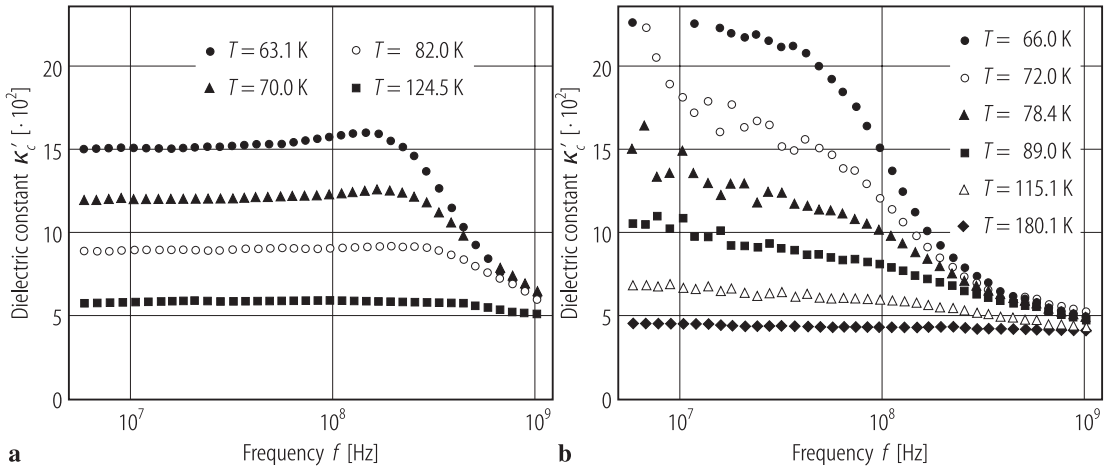

Fig. 1A-10-253. $BaTiO_3$ (hexagonal). κ_c' vs. f [88Deg]. Parameter: T. (**a**) after zero field cooling. (**b**) after field cooling of $E_{bias} = 1\ MV\,m^{-1}$ from 210 K to 74 K.

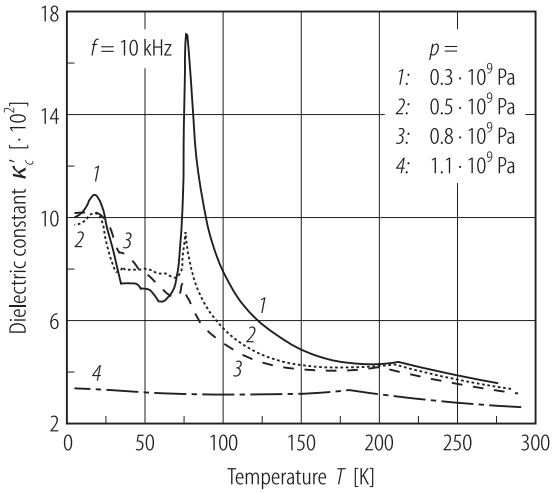

Fig. 1A-10-254. $BaTiO_3$ (hexagonal). κ_c' vs. T [94Aki2].
Parameter: p, hydrostatic pressure. f = 10 kHz.

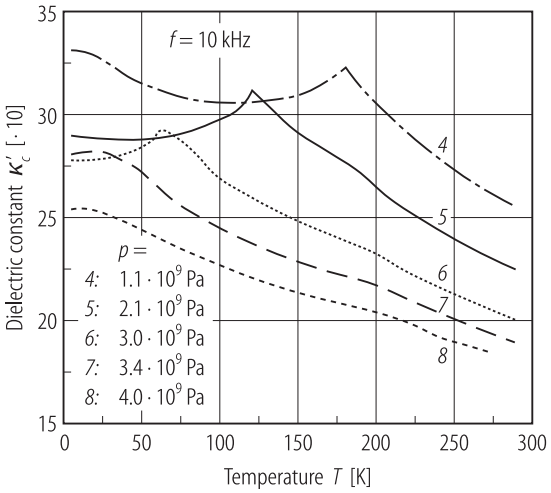

Fig. 1A-10-255. $BaTiO_3$ (hexagonal). κ_c' vs. T [94Aki2]. Parameter: p, hydrostatic pressure. f = 10 kHz.

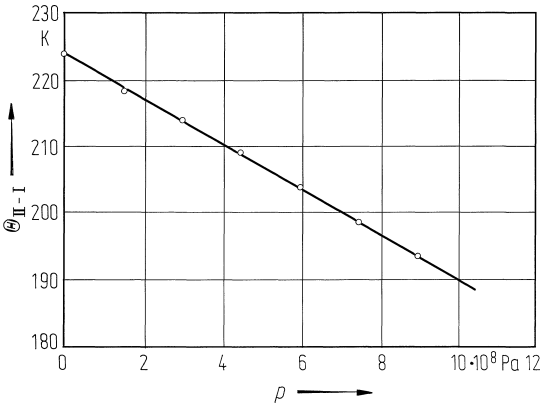

Fig. 1A-10-256. $BaTiO_3$ (hexagonal, ceramics). Θ_{II-I} vs. p [88Aki1]. p: hydrostatic pressure.

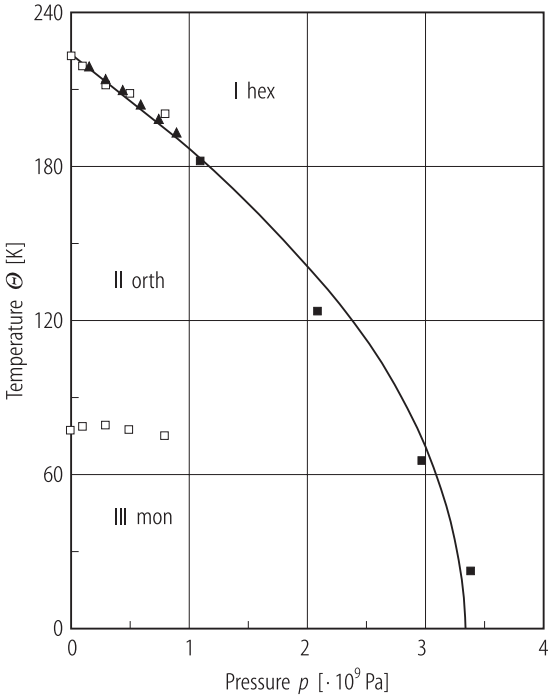

Fig. 1A-10-257. $BaTiO_3$ (hexagonal). Θ vs. p [94Aki2]. p: hydrostatic pressure. Open and full squares: single crystals. Full triangle: ceramics (see Fig. 1A-10-256). Solid line: calculated result with $\Theta_{II-I} = a\ (p_c - p)^{1/2}$ where critical pressure $p_c = 3.4 \cdot 10^9$ Pa.

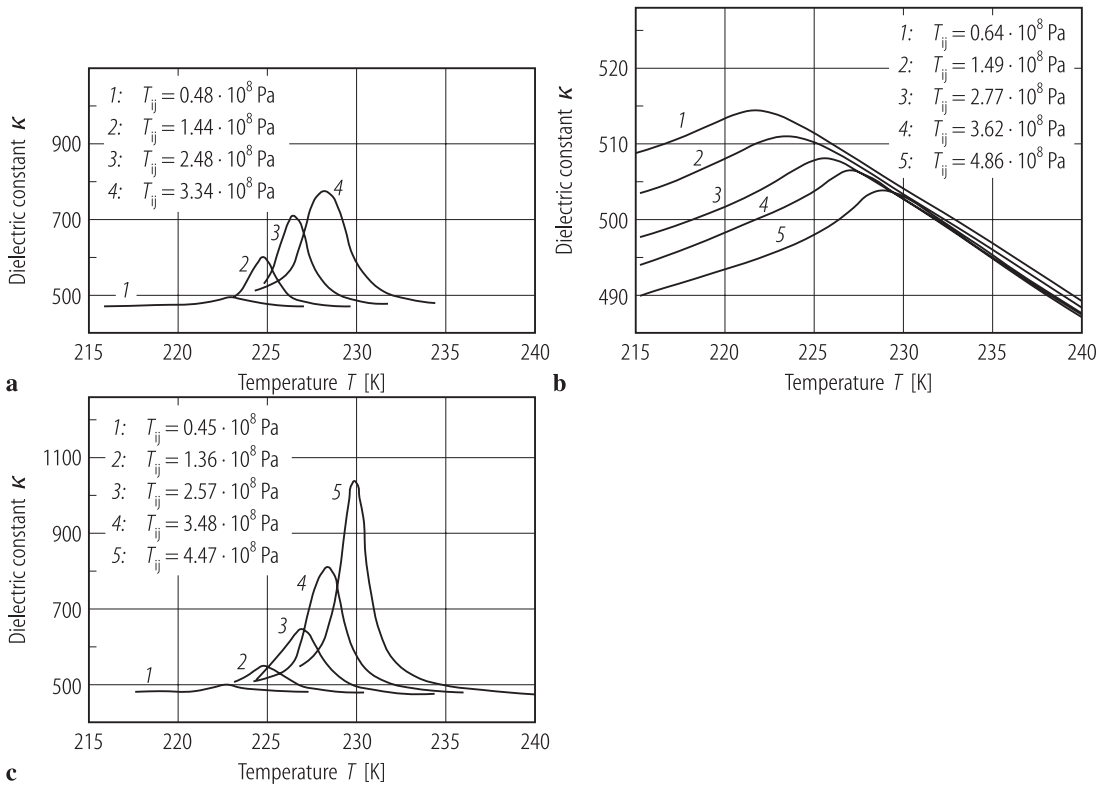

Fig. 1A-10-258. $BaTiO_3$ (hexagonal). κ vs. T_{ij} [91Yam]. Parameter: T_{ij}, uniaxial stress. f = 10 kHz. (**a**) T_{ij} || [100]. (**b**) T_{ij} || [010]. (**c**) T_{ij} || [110].

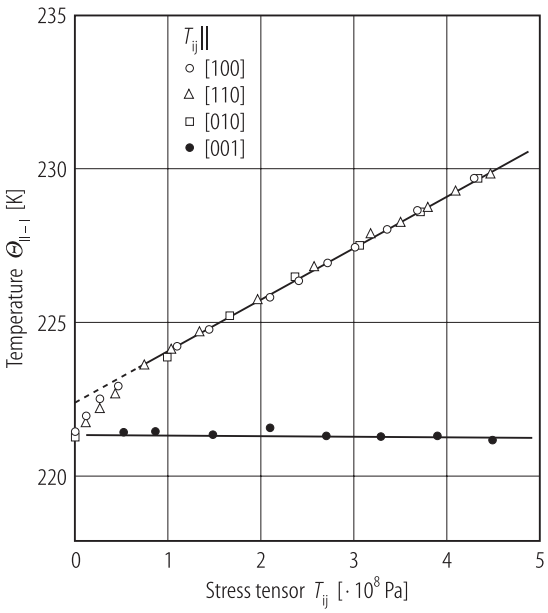

Fig. 1A-10-259. $BaTiO_3$ (hexagonal). $\Theta_{II–I}$ vs. T_{ij} [91Yam]. T_{ij}: uniaxial stress.

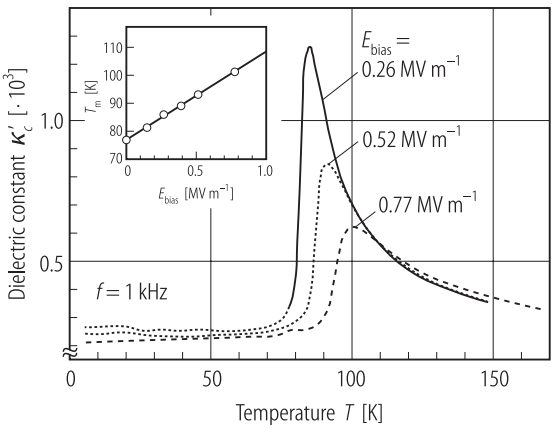

Fig. 1A-10-260. $BaTiO_3$ (hexagonal). κ_c' vs. T [89Aki]. Parameter: E_{bias}. The insert shows T_m vs. E_{bias}. T_m: temperature for κ_c' peak. f = 1 kHz.

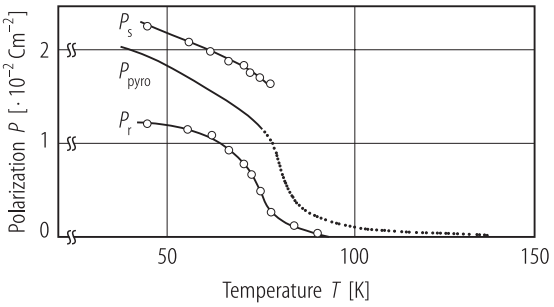

Fig. 1A-10-261. $BaTiO_3$ (hexagonal). P_s, P_r, P_{pyro} vs. T [89Aki]. P_{pyro}: pyroelectric charge. P_s and P_r were obtained from D–E hysteresis curves at $8.3 \cdot 10^{-3}$ Hz.

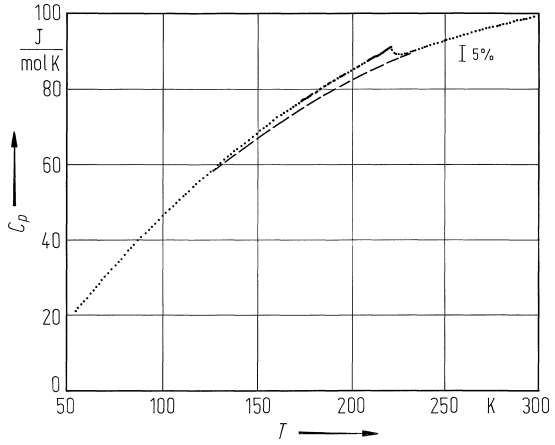

Fig. 1A-10-262. $BaTiO_3$ (hexagonal, ceramics). C_p vs. T [88Aki3]. C_p: molar heat capacity at constant pressure. The broken line represents the normal portion of heat capacity given by $C_p(\text{base}) = A + BT + CT^2 + DT^3$, where A, B, C and D are constants.

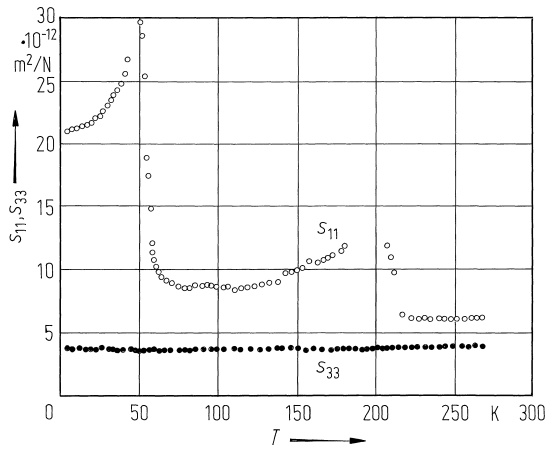

Fig. 1A-10-263. $BaTiO_3$ (hexagonal). s_{11}, s_{33} vs. T [88Yam2]. s_{11}, s_{33}: elastic compliances.

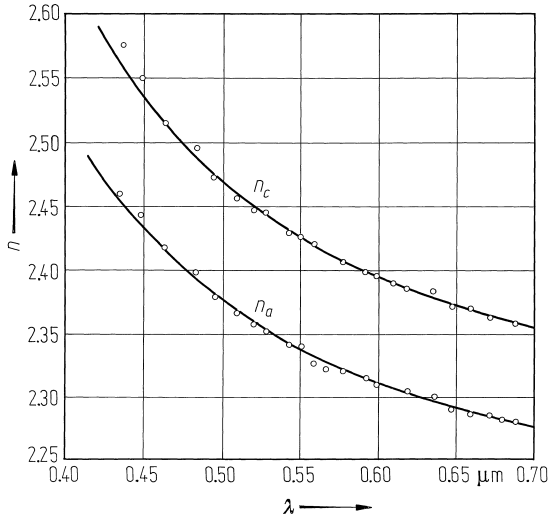

Fig. 1A-10-264. $BaTiO_3$ (hexagonal). n_a, n_c vs. λ [86Aki].
T = RT.

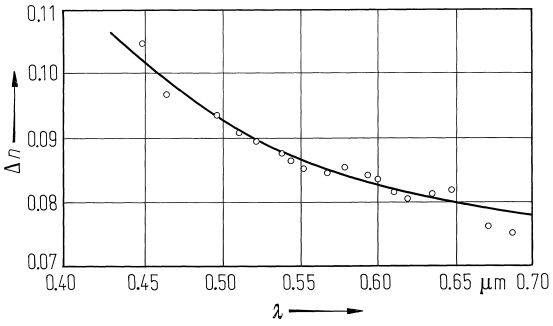

Fig. 1A-10-265. $BaTiO_3$ (hexagonal). Δn vs. λ [86Aki]. $\Delta n = n_c - n_a$. T = RT. Solid line is the fit of the data to single term Sellmeier oscillator model.

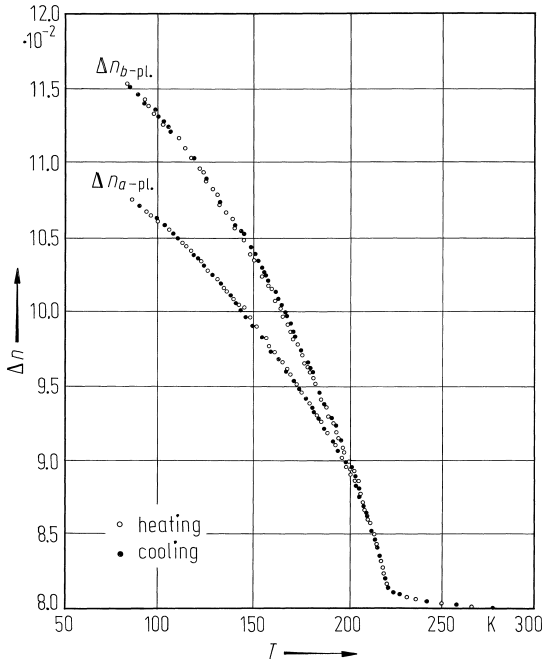

Fig. 1A-10-266. $BaTiO_3$ (hexagonal). Δn vs. T [86Aki]. $\Delta n_{a-pl.}$ and $\Delta n_{b-pl.}$ are birefringence measured for the a and b plate, respectively. λ = 633 nm.

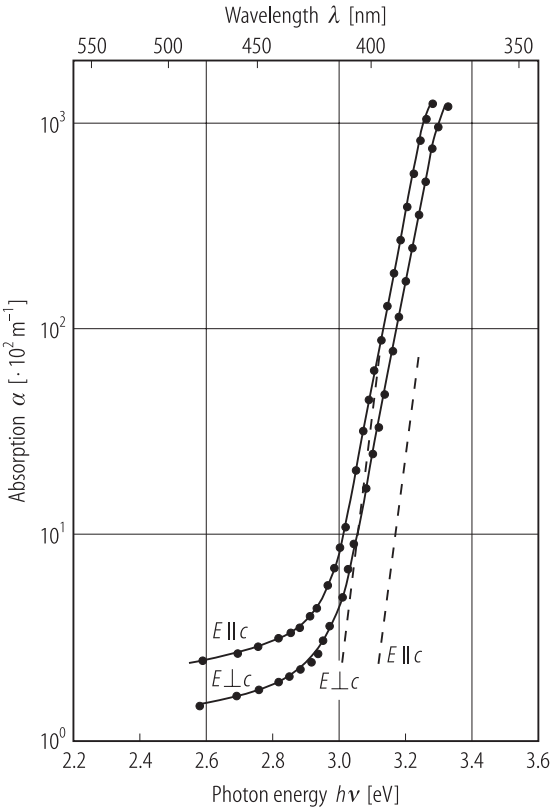

Fig. 1A-10-267. $BaTiO_3$ (hexagonal). α vs. $h\nu$ at RT [95Aki1]. α: optical absorption coefficient. Solid curves: hexagonal, broken curves: tetragonal [70Wem].

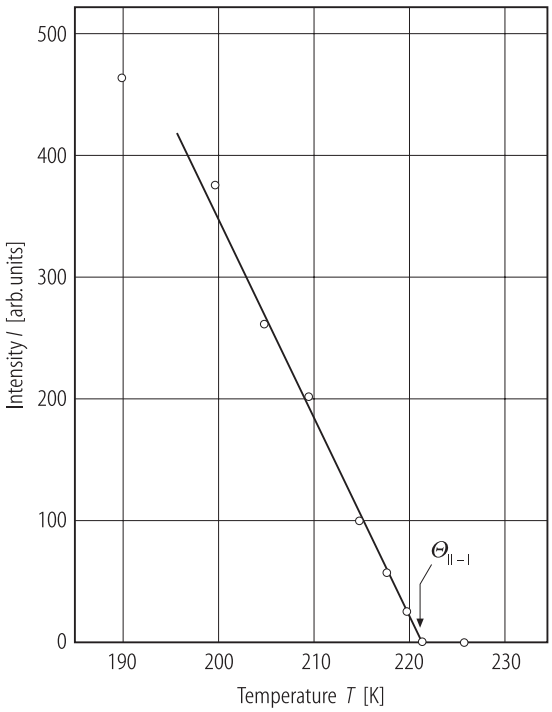

Fig. 1A-10-268. $BaTiO_3$ (hexagonal). I vs. T [88Ino].
I: relative intensity of optical second harmonic generation.

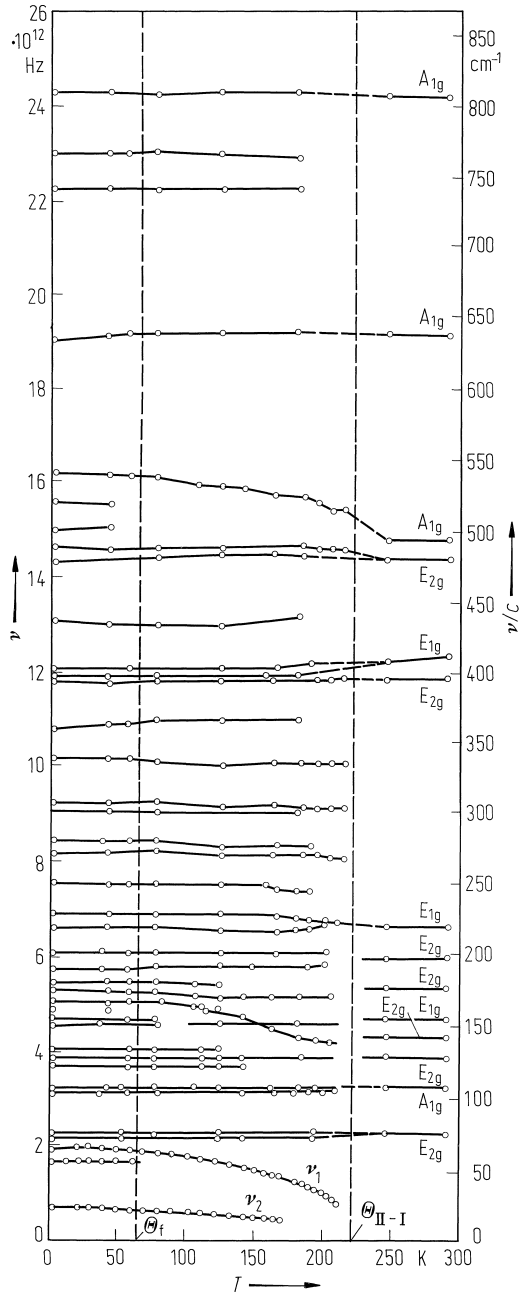

Fig. 1A-10-269. $BaTiO_3$ (hexagonal). ν vs. T [87Yam]. ν: the frequencies of Raman peaks. A_{1g} etc.: assignment of Raman active phonon modes on the basis of the space group of $P6_3/mmc$ with six formula units in a unit cell. ν_1 and ν_2: the frequencies of two lowest soft phonon modes.

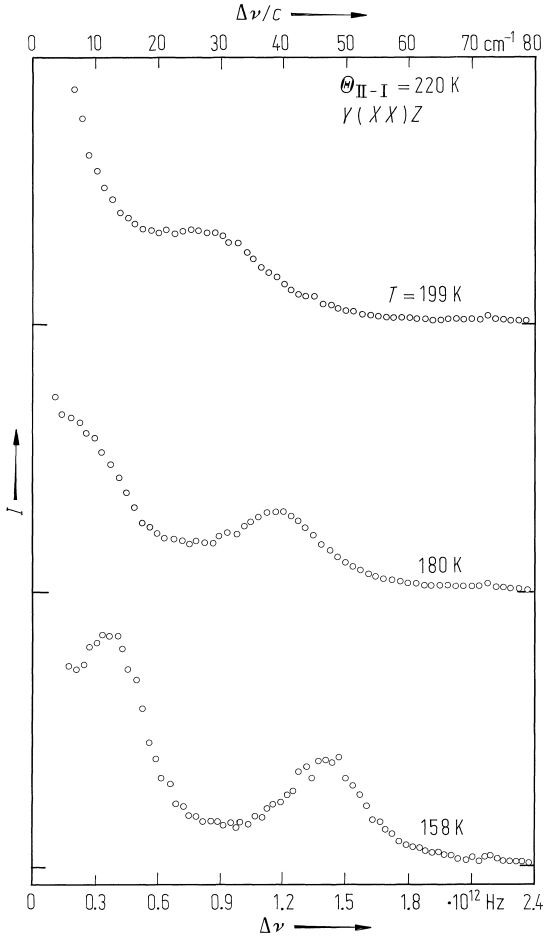

Fig. 1A-10-270. $BaTiO_3$ (hexagonal). I vs. $\Delta\nu$ [87Yam]. I: intensity of Raman scattering from the soft-phonon modes in phase II associated with the structural phase transition at $\Theta_{II\text{-}I}$. $\Delta\nu$: frequency shift. Parameter: T.

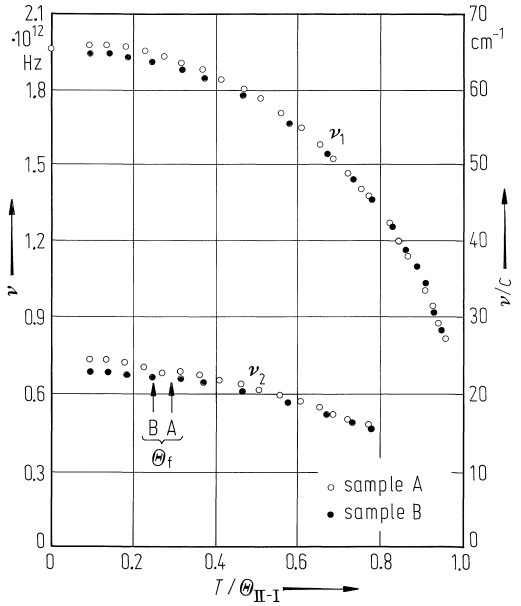

Fig. 1A-10-271. $BaTiO_3$ (hexagonal). ν_1, ν_2 vs. $T/\Theta_{II–I}$ [87Yam]. ν_1, ν_2: the frequencies of two lowest soft phonon modes.

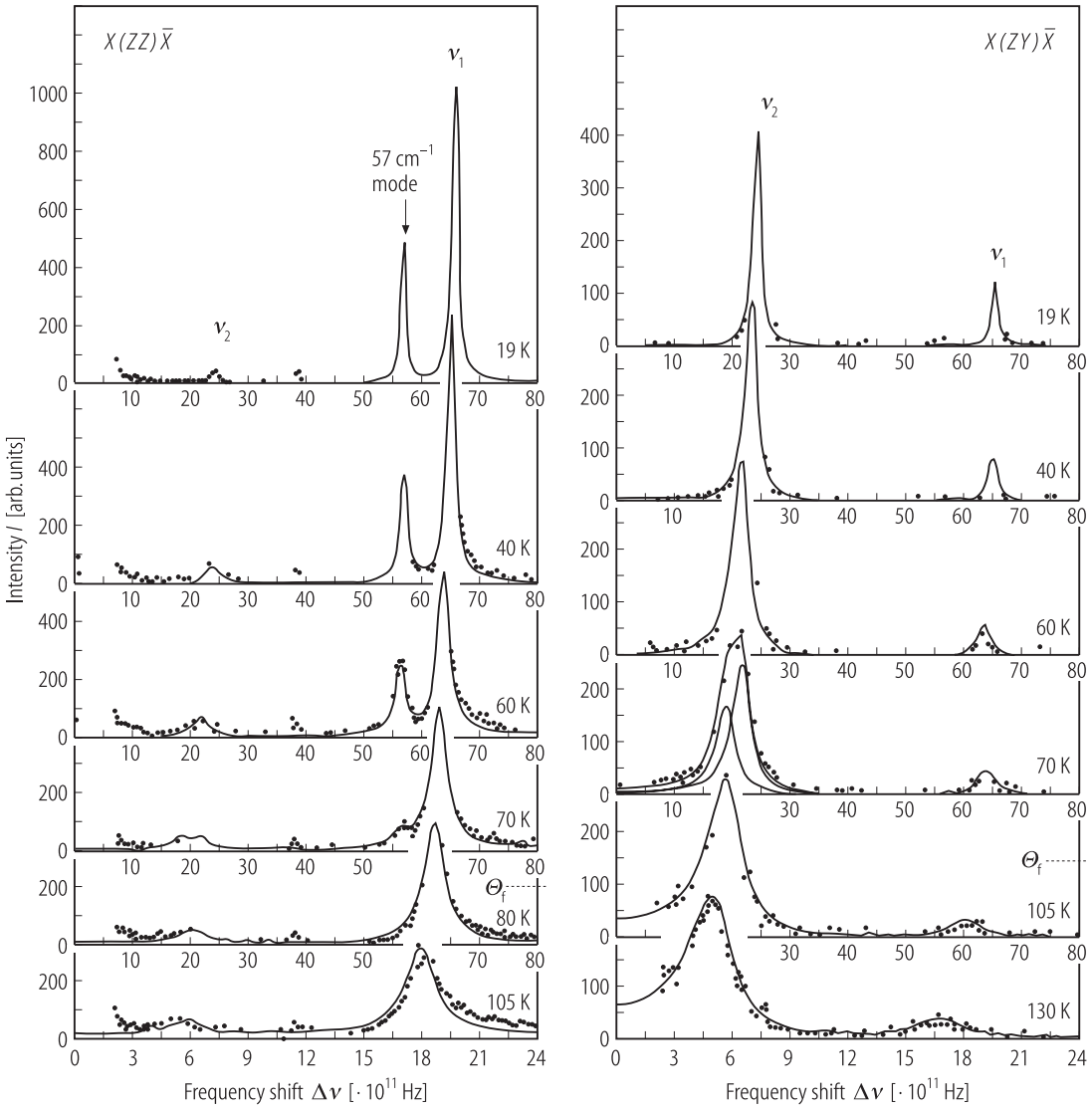

Fig. 1A-10-272. $BaTiO_3$ (hexagonal). I vs. $\Delta\nu$ [94Aki3]. I: Raman scattering intensity of $X(ZZ)\,\overline{X}$ and $X(ZY)\,\overline{X}$ geometric arrangement around Θ_f. $\Delta\nu$: frequency shift. Parameter: T.

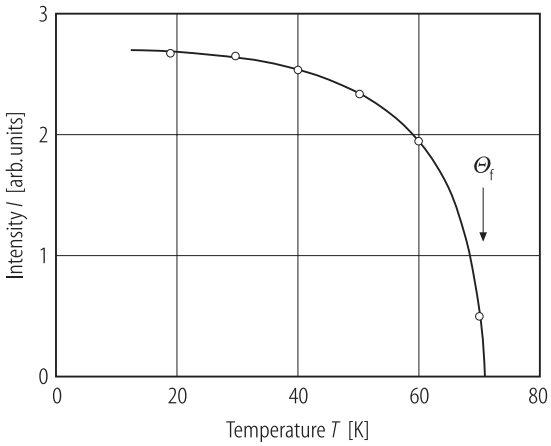

Fig. 1A-10-273. $BaTiO_3$ (hexagonal). I vs. T [94Aki3].
I: Raman scattering intensity of a peak at 57 cm^{-1}.

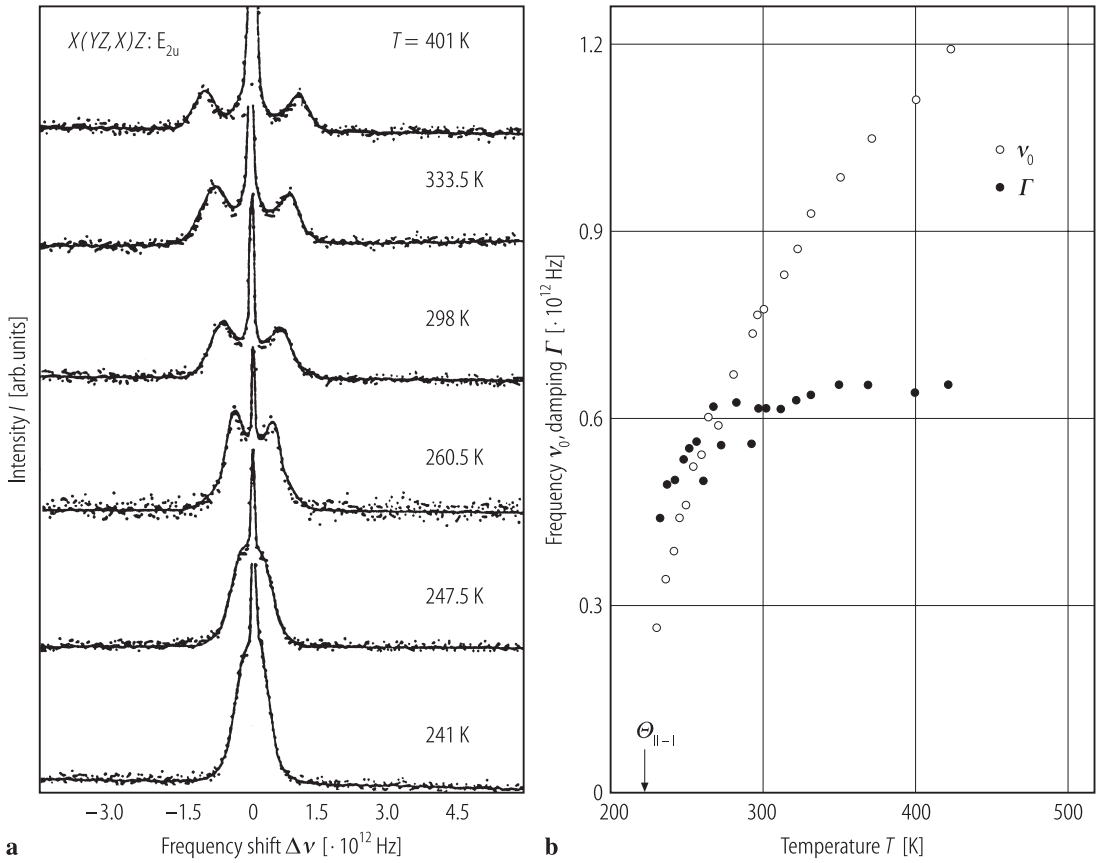

Fig. 1A-10-274. $BaTiO_3$ (hexagonal). Temperature dependence of E_{2u} soft phonon in the phase I [88Ino]. (**a**): I vs. $\Delta\nu$. I: hyper-Raman scattering intensity. $\Delta\nu$: frequency shift. Parameter: T. (**b**): ν_o and Γ vs. T. ν_o and Γ: the frequency and the damping constant of the E_{2u} soft phonon, respectively.

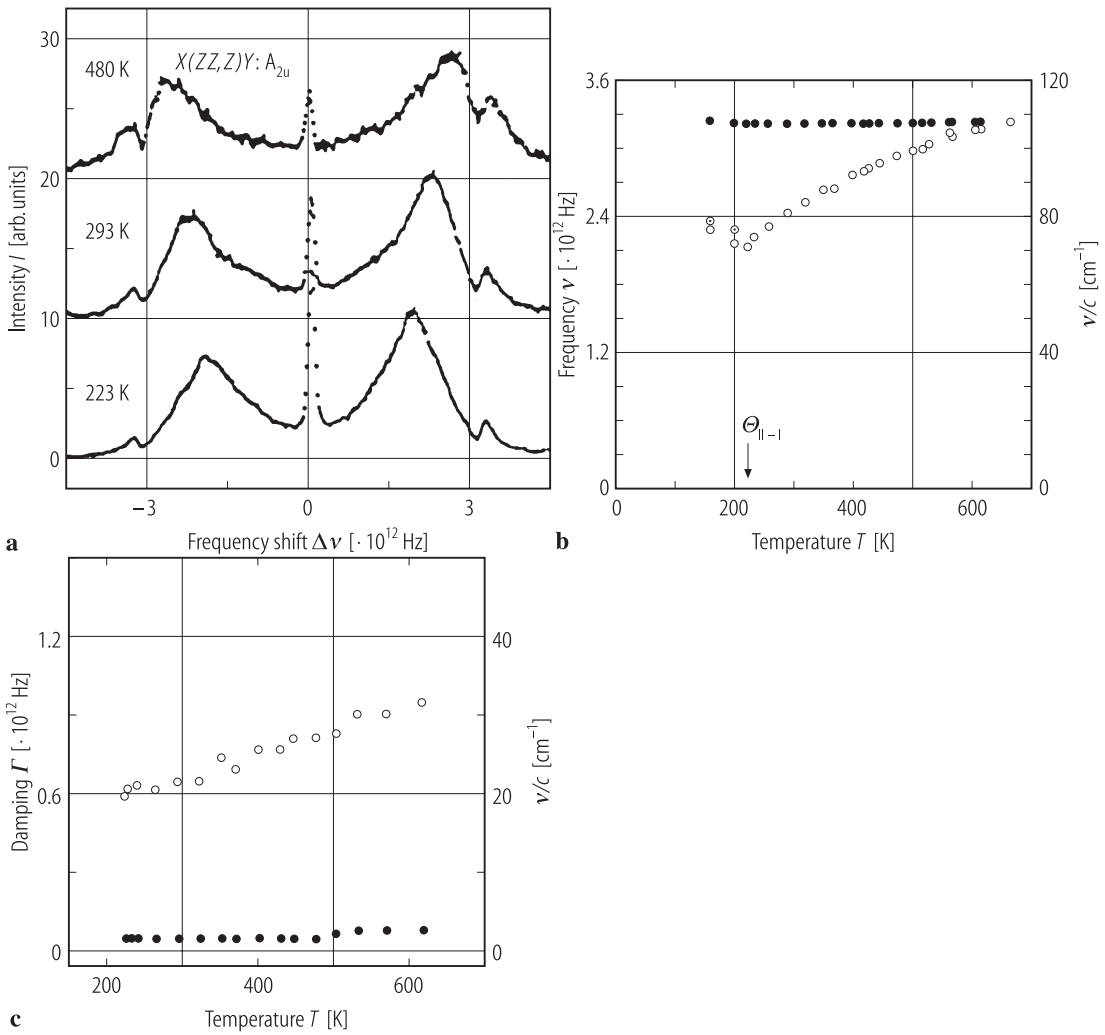

Fig. 1A-10-275. $BaTiO_3$ (hexagonal). Temperature dependence of the low frequency A_{2u} phonons [94Yam1]. (**a**): I vs. $\Delta\nu$. I: hyper-Raman scattering intensity. $\Delta\nu$: frequency shift. Parameter: T. (**b**): ν vs. T. ν: mode frequencies. Open circles: soft-mode, full circles: second-lowest frequency mode. (**c**): Γ vs. T. Γ: damping constants.

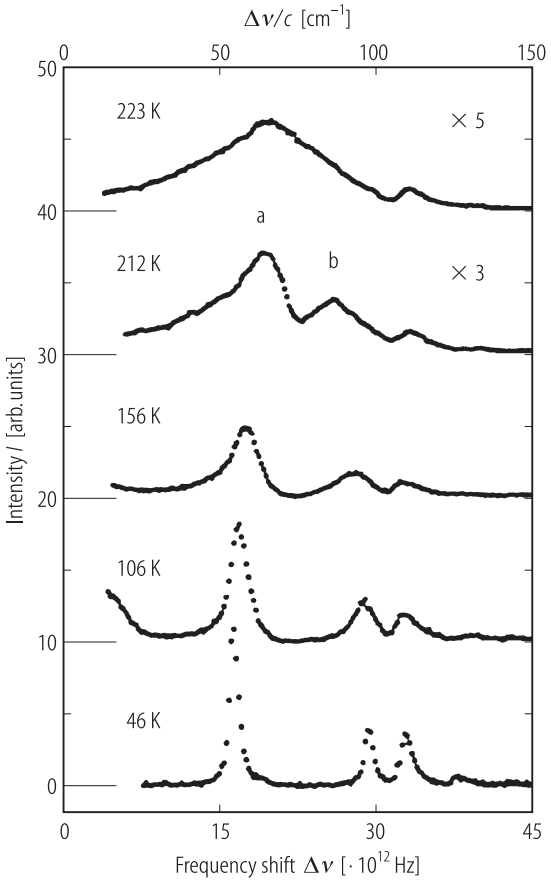

Fig. 1A-10-276. $BaTiO_3$ (hexagonal). *I* vs. $\Delta\nu$ [96Yam1]. *I*: hyper-Raman scattering intensity observed in the *X*(*ZZ*,*Z*)*Y* geometric arrangement. $\Delta\nu$: frequency shift. Parameter: *T*.

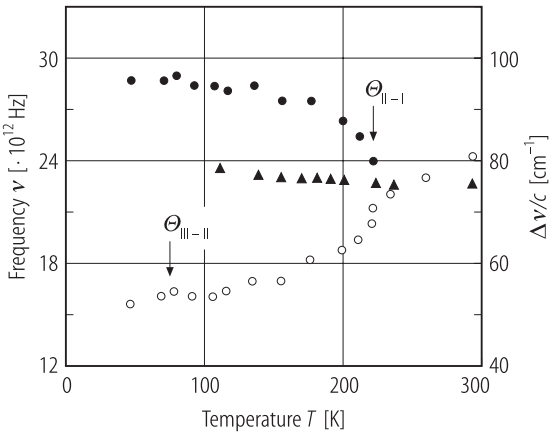

Fig. 1A-10-277. $BaTiO_3$ (hexagonal). ν vs. T [96Yam]. ν: phonon mode frequency observed in the hyper-Raman scattering in the $X(ZZ,Z)Y$ geometric arrangement.

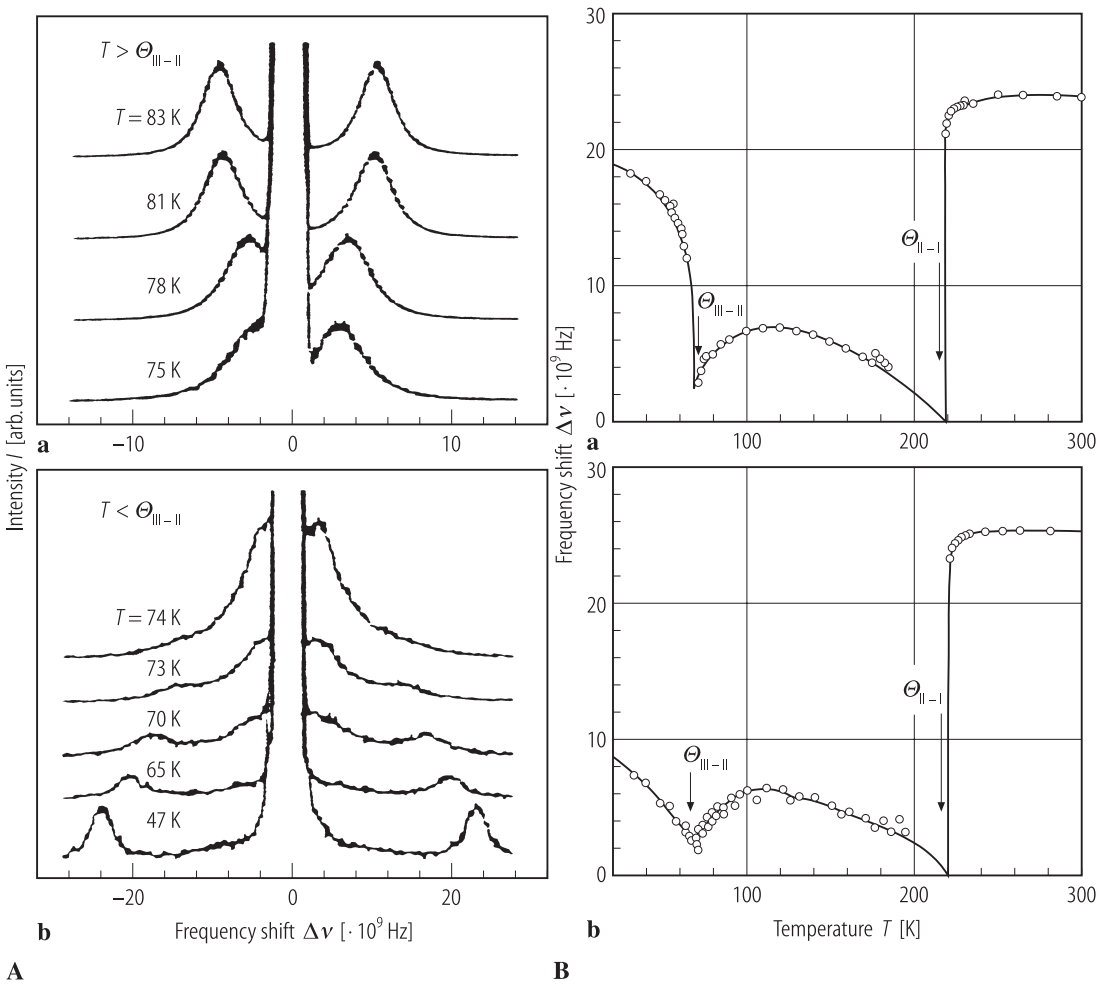

Fig. 1A-10-278. $BaTiO_3$ (hexagonal). Temperature dependence of Brillouin scattering spectra [95Yam]. (A): Brillouin scattering intensity vs. T around $\Theta_{III–II}$. $\Delta\nu$: frequency shift. Parameter: T. (B): Brillouin scattering frequency shift of the c_{66} mode observed without uniaxial stress (**a**) and under uniaxial stress (**b**).

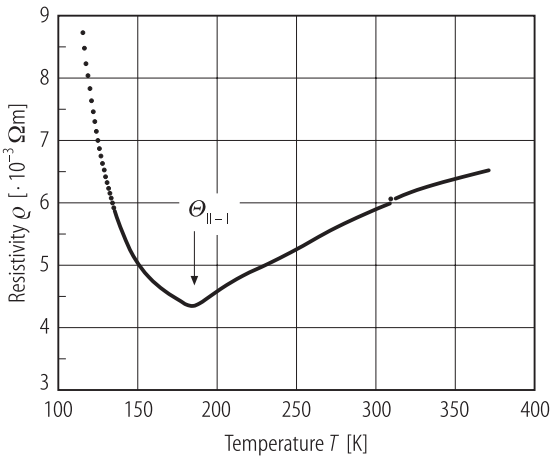

Fig. 1A-10-279. $BaTiO_3$ (hexagonal, reduced). ρ vs. T [95Aki2].

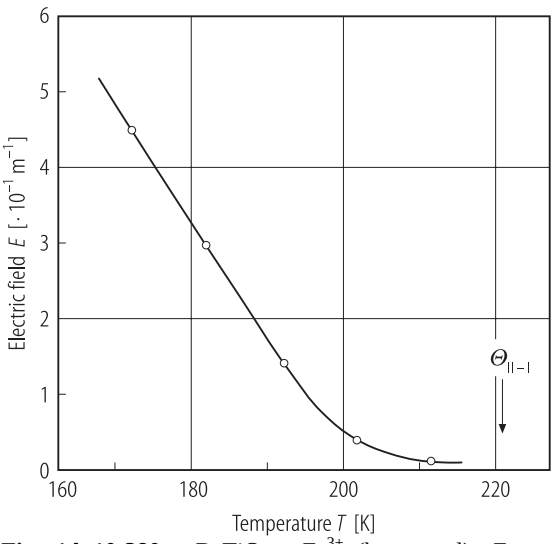

Fig. 1A-10-280. $BaTiO_3$: Fe^{3+} (hexagonal). E vs. T [89Ohi]. E: spin Hamiltonian parameter of Fe^{3+}.

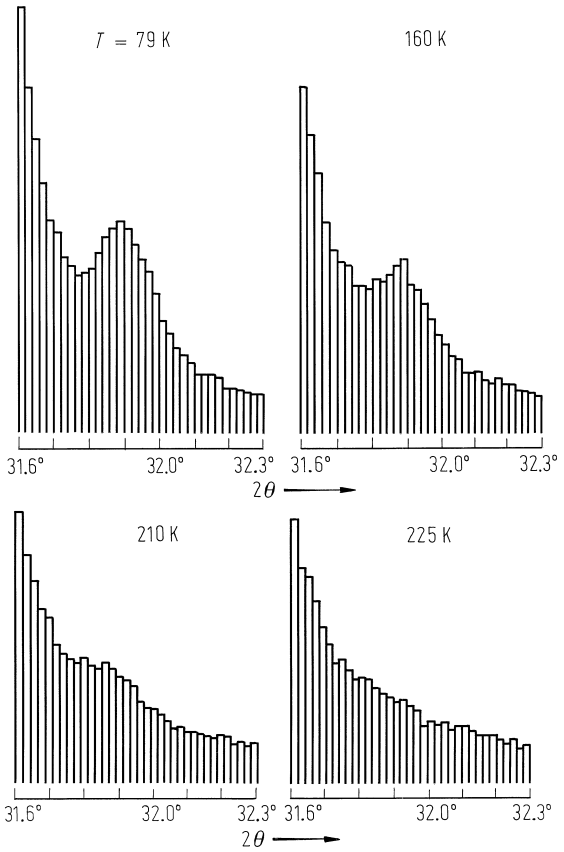

Fig. 1A-10-281. $BaTiO_3$ (hexagonal). X-ray diffraction profiles of hexagonal $BaTiO_3$ powder around $2\theta = 32^o$ [85Saw2]. Parameter: *T*. A small knoll at $2\theta = 31.9^o$ at 210 K grows into a peak at low temperature.

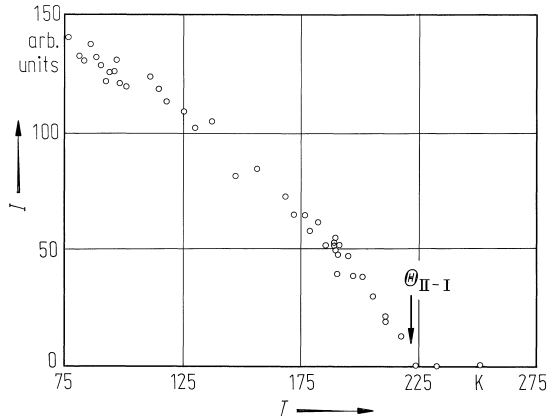

Fig. 1A-10-282. $BaTiO_3$ (hexagonal). I vs. T [85Saw2].
I: integrated intensity of a diffraction peak at $2\theta = 31.9^o$.
See also Fig. 1A-10-281.

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