Unraveling Relaxor Phase Transitions by k-Space Spectroscopy

KLAUS BETZLER, CHRISTOPH GÖDEKER, URS HEINE, UWE VOELKER

UNIVERSITÄT OSNABRÜCK

2009 WILLIAMSBURG WORKSHOP ON
FUNDAMENTAL PHYSICS OF FERROELECTRICS

Relaxor Ferroelectrics

G. A. Smolenskii 1954: Segnetoelektricheskie svoistva tverdykh rastvorov stannata bariya v titanate bariya

G. A. Smolenskii 1958: Dielectric polarization and losses of some complex compounds

Many Others: . . .

. . .

L. Eric Cross 1987: Relaxor ferroelectrics

Strontium Barium Niobate Revisited

Eur. Phys. J. B **14**, 633–637 (2000)

Phase transitions in $Sr_{0.61}Ba_{0.39}Nb_2O_6$:$Ce^{3+}$: II. Linear birefringence studies of spontaneous and precursor polarization

P. Lehnen[1], W. Kleemann[1,a], Th. Woike[2], and R. Pankrath[3]

Strontium Barium Niobate Revisited

Eur. Phys. J. B 14, 633–637 (2000)

Phase transitions in $Sr_{0.61}Ba_{0.39}Nb_2O_6:Ce^{3+}$:
II. Linear birefringence studies of spontaneous and precursor polarization

P. Lehnen[1], W. Kleemann[1,a], Th. Woike[2], and R. Pankrath[3]

PHYSICAL REVIEW B, VOLUME 64, 134109 (2001)

^{93}Nb NMR of the random-field-dominated relaxor transition in pure and doped SBN

R. Blinc, A. Gregorovič, B. Zalar, R. Pirc, and J. Seliger W. Kleemann S. G. Lushnikov R. Pankrath

Strontium Barium Niobate Revisited

Eur. Phys. J. B 14, 633–637 (2000)

Phase transitions in $Sr_{0.61}Ba_{0.39}Nb_2O_6:Ce^{3+}$: II. Linear birefringence studies of spontaneous and precursor polarization

P. Lehnen[1], W. Kleemann[1,a], Th. Woike[2], and R. Pankrath[3]

PHYSICAL REVIEW B, VOLUME 64, 134109 (2001)

^{93}Nb NMR of the random-field-dominated relaxor transition in pure and doped SBN

R. Blinc, A. Gregorovič, B. Zalar, R. Pirc, and J. Seliger W. Kleemann S. G. Lushnikov R. Pankrath

VOLUME 86, NUMBER 26 PHYSICAL REVIEW LETTERS 25 JUNE 2001

Dynamic Light Scattering at Domains and Nanoclusters in a Relaxor Ferroelectric

W. Kleemann,[1] P. Licinio,[2] Th. Woike,[3] and R. Pankrath[4]

Strontium Barium Niobate Revisited

Eur. Phys. J. B 14, 633–637 (2000)

**Phase transitions in $Sr_{0.61}Ba_{0.39}Nb_2O_6:Ce^{3+}$:
II. Linear birefringence studies of spontaneous and precursor polarization**

P. Lehnen[1], W. Kleemann[1,a], Th. Woike[2], and R. Pankrath[3]

PHYSICAL REVIEW B, VOLUME 64, 134109 (2001)

^{93}Nb NMR of the random-field-dominated relaxor transition in pure and doped SBN

R. Blinc, A. Gregorovič, B. Zalar, R. Pirc, and J. Seliger W. Kleemann S. G. Lushnikov R. Pankrath

VOLUME 86, NUMBER 26 PHYSICAL REVIEW LETTERS 25 JUNE 2001

Dynamic Light Scattering at Domains and Nanoclusters in a Relaxor Ferroelectric

W. Kleemann,[1] P. Licinio,[2] Th. Woike,[3] and R. Pankrath[4]

PHYSICAL REVIEW B, VOLUME 64, 224109 (2001)

Ferroelectric nanodomains in the uniaxial relaxor system $Sr_{0.61-x}Ba_{0.39}Nb_2O_6:Ce_x^{3+}$

P. Lehnen and W. Kleemann Th. Woike R. Pankrath

Strontium Barium Niobate Revisited

Eur. Phys. J. B 14, 633–637 (2000)

Phase transitions in $Sr_{0.61}Ba_{0.39}Nb_2O_6:Ce^{3+}$:
II. Linear birefringence studies of spontaneous and precursor polarization

P. Lehnen, W. Kleemann, Th. Woike, and R. Pankrath[3]

Europhys. Lett., 57 (1), pp. 14–19 (2002)
1 January 2002

Uniaxial relaxor ferroelectrics: The ferroic random-field Ising model materialized at last

W. Kleemann[1], J. Dec[1](*), P. Lehnen[1], R. Blinc[2], B. Zalar[2] and R. Pankrath[3]

PHYSICAL REVIEW B, VOLUME 64, 134109 (2001)

^{93}Nb NMR of the random-field-dominated relaxor transition in pure and doped SBN

R. Blinc, A. Gregorovič, B. Zalar, R. Pirc, and J. Seliger W. Kleemann S. G. Lushnikov R. Pankrath

VOLUME 86, NUMBER 26 PHYSICAL REVIEW LETTERS 25 JUNE 2001

Dynamic Light Scattering at Domains and Nanoclusters in a Relaxor Ferroelectric

W. Kleemann,[1] P. Licinio,[2] Th. Woike,[3] and R. Pankrath[4]

PHYSICAL REVIEW B, VOLUME 64, 224109 (2001)

Ferroelectric nanodomains in the uniaxial relaxor system $Sr_{0.61-x}Ba_{0.39}Nb_2O_6:Ce_x^{3+}$

P. Lehnen and W. Kleemann Th. Woike R. Pankrath

Strontium Barium Niobate Revisited

Europhys. Lett., 57 (1), pp. 14–19 (2002)

1 January 2002

Uniaxial relaxor ferroelectrics: The ferroic random-field Ising model materialized at last

W. Kleemann[1], J. Dec[1](*), P. Lehnen[1], R. Blinc[2], B. Zalar[2] and R. Pankrath[3]

Volume 92, Number 6 — PHYSICAL REVIEW LETTERS — week ending 13 FEBRUARY 2004

Change from 3D-Ising to Random Field-Ising-Model Criticality in a Uniaxial Relaxor Ferroelectric

T. Granzow and Th. Woike — M. Wöhlecke and M. Imlau — W. Kleemann

Eur. Phys. J. B 14, 633–637 (2000)

Phase transitions in $Sr_{0.61}Ba_{0.39}Nb_2O_6:Ce^{3+}$: II. Linear birefringence studies of spontaneous and precursor polarization

P. Lehnen[1], W. Kleemann[1], Th. Woike[2], and R. Pankrath[3]

PHYSICAL REVIEW B, VOLUME 64, 134[…] (2001)

^{93}Nb NMR of the random-field-dominated relaxor transition in pure and doped SBN

R. Blinc, A. Gregorovič, B. Zalar, R. Pirc, and J. Seliger — W. Kleemann — S. G. Lushnikov — R. Pankrath

VOLUME 86, NUMBER 26 — PHYSICAL REVIEW LETTERS — 25 JUNE 2001

Dynamic Light Scattering at Domains and Nanoclusters in a Relaxor Ferroelectric

W. Kleemann,[1] P. Licinio,[2] Th. Woike,[3] and R. Pankrath[4]

PHYSICAL REVIEW B, VOLUME 64, 224109 (2001)

Ferroelectric nanodomains in the uniaxial relaxor system $Sr_{0.61-x}Ba_{0.39}Nb_2O_6:Ce_x^{3+}$

P. Lehnen and W. Kleemann — Th. Woike — R. Pankrath

Strontium Barium Niobate Revisited

Europhys. Lett., 57 (1), pp. 14–19 (2002) — 1 January 2002

Uniaxial relaxor ferroelectrics: The ferroic random-field Ising model materialized at last

W. Kleemann[1], J. Dec[1](*), P. Lehnen[1], R. Blinc[2], B. Zalar[2] and R. Pankrath[3]

Volume 92, Number 6 — PHYSICAL REVIEW LETTERS — week ending 13 FEBRUARY 2004

Change from 3D-Ising to Random Field-Ising-Model Criticality in a Uniaxial Relaxor Ferroelectric

W. Kleemann, T. Granzow and Th. Woike, M. Wöhlecke and M. Imlau

PRL 97, 065702 (2006) — PHYSICAL REVIEW LETTERS — week ending 11 AUGUST 2006

Two-Dimensional Ising Model Criticality in a Three-Dimensional Uniaxial Relaxor Ferroelectric with Frozen Polar Nanoregions

Wolfgang Kleemann,[1,*] Jan Dec,[1,2] Vladimir V. Shvartsman,[1] Zdravko Kutnjak,[3] and Thomas Braun[1]

Eur. Phys. J. B 14, 633–637 (2000)

Phase transitions in $Sr_{0.61}Ba_{0.39}Nb_2O_6:Ce^{3+}$: II. Linear birefringence studies of spontaneous and precursor polarization

P. Lehnen[1], W. Kleemann[1], Th. Woike[2], and R. Pankrath[3]

PHYSICAL REVIEW B, VOLUME 64, 134104 (2001)

^{93}Nb NMR of the random-field-dominated relaxor transition in pure and doped SBN

R. Blinc, A. Gregorovič, B. Zalar, R. Pirc and J. Seliger; W. Kleemann; S. G. Lushnikov; R. Pankrath

VOLUME 86, NUMBER 26 — PHYSICAL REVIEW LETTERS — 25 JUNE 2001

Dynamic Light Scattering at Domains and Nanoclusters in a Relaxor Ferroelectric

W. Kleemann,[1] P. Licinio,[2] Th. Woike,[3] and R. Pankrath[4]

PHYSICAL REVIEW B, VOLUME 64, 224109 (2001)

Ferroelectric nanodomains in the uniaxial relaxor system $Sr_{0.61-x}Ba_{0.39}Nb_2O_6:Ce_x^{3+}$

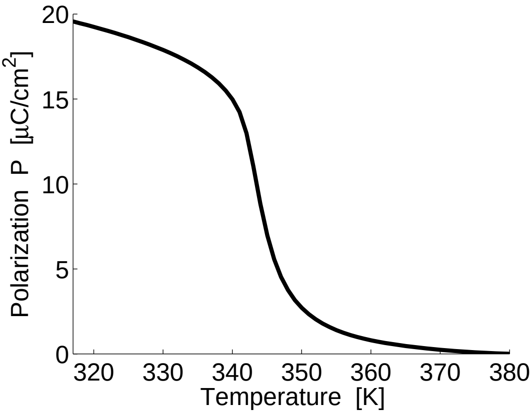
P. Lehnen and W. Kleemann; Th. Woike; R. Pankrath

Critical Exponents ? $\Longrightarrow$ $\beta = 0.1 \ldots 0.35$?

Order parameter $P(T) = P_0 \left(1 - \frac{T}{T_C}\right)^{\beta}$ for $T \lesssim T_C$

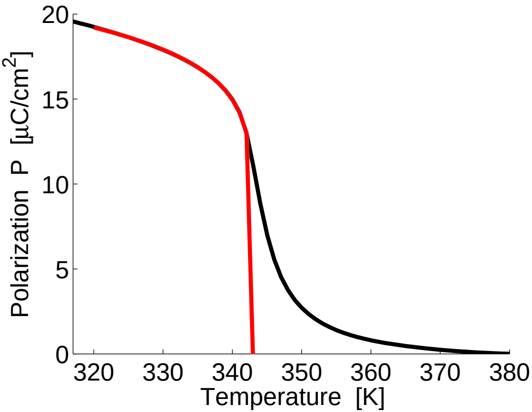
Critical Exponents ? $\Longrightarrow$ $\beta = 0.1 \ldots 0.35$?

Order parameter $P(T) = P_0 \left(1 - \frac{T}{T_C}\right)^{\beta}$ for $T \lessapprox T_C$



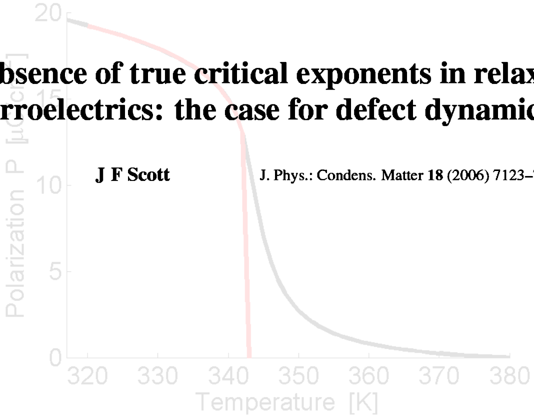
Critical Exponents ? $\Longrightarrow$ $\beta = 0.1 \ldots 0.35$?

Order parameter $P(T) = P_0 \left(1 - \frac{T}{T_C}\right)^{\beta}$ for $T \lessapprox T_C$



Critical Exponents ? $\Longrightarrow$ $\beta = 0.1 \ldots 0.35$?

Order parameter $P(T) = P_0 \left(1 - \frac{T}{T_C}\right)^{\beta}$ for $T \lessapprox T_C$



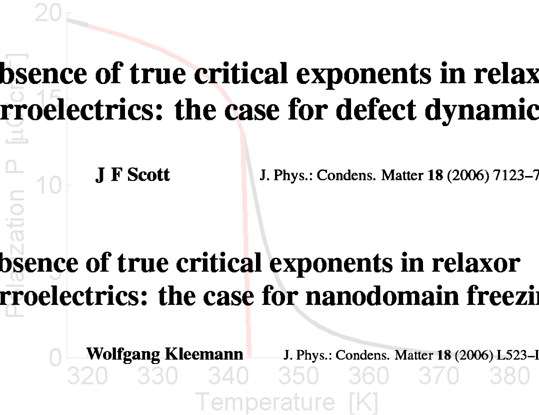
Absence of true critical exponents in relaxor ferroelectrics: the case for defect dynamics

J F Scott

J. Phys.: Condens. Matter **18** (2006) 7123–7134

Critical Exponents ? $\Longrightarrow$ $\beta = 0.1 \ldots 0.35$?

Order parameter $P(T) = P_0 \left(1 - \frac{T}{T_C}\right)^{\beta}$ for $T \lessapprox T_C$



Absence of true critical exponents in relaxor ferroelectrics: the case for defect dynamics

J F Scott J. Phys.: Condens. Matter **18** (2006) 7123–7134

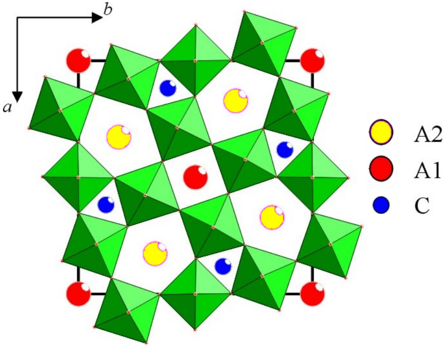
Absence of true critical exponents in relaxor ferroelectrics: the case for nanodomain freezing

Wolfgang Kleemann J. Phys.: Condens. Matter **18** (2006) L523–L526

Outline

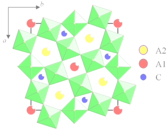
Strontium Barium Niobate – SBN – $Sr_xBa_{1-x}Nb_2O_6$

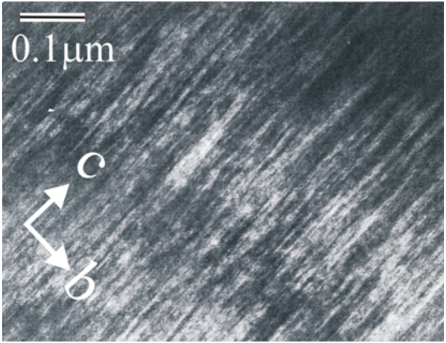
SBN – $Sr_xBa_{1-x}Nb_2O_6$ – Structure



Sergey Podlozhenov, Heribert A. Graetsch, Julius Schneider, Michael Ulex, Manfred Wöhlecke and Klaus Betzler: *Crystal structure of strontium barium niobate $Sr_xBa_{1-x}Nb_2O_6$ (SBN) in the composition range $0.32 < x < 0.82$*. Acta Cryst. B 62:960–965 (2006).

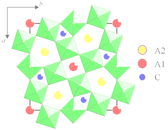
SBN – $Sr_xBa_{1-x}Nb_2O_6$ – Structure

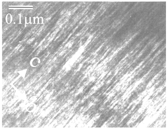


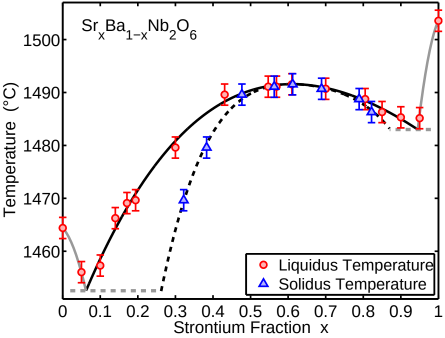


D. Viehland, Z. Xu, W.-H. Huang: *Structure-property relationships in strontium barium niobate. 1. needle-like nanopolar domains and the metastably-locked incommensurate structure.* Phil. Mag. A 71:205 (1995)

SBN – $Sr_xBa_{1-x}Nb_2O_6$ – Phase Diagramm

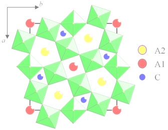


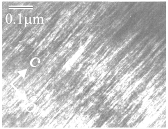


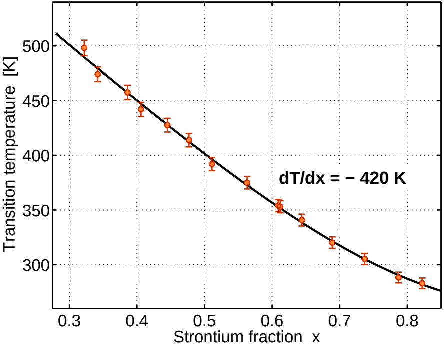


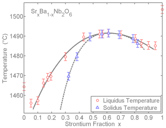
Michael Ulex, Rainer Pankrath, Klaus Betzler: *Growth of strontium barium niobate: the liquidus-solidus phase diagram*. J. Crystal Growth 271:128–133 (2004).

SBN – $Sr_xBa_{1-x}Nb_2O_6$ – Transition Temperature



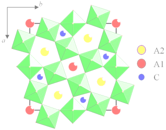


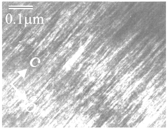


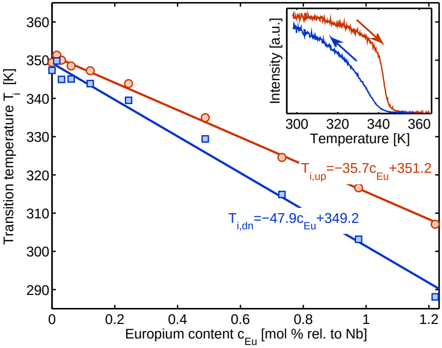


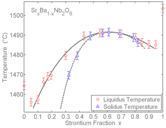
C. David, T. Granzow, A. Tunyagi, M. Wöhlecke, Th. Woike, K. Betzler, M. Ulex, M. Imlau, R. Pankrath: *Composition dependence of the phase transition temperature in Strontium Barium Niobate.* phys. stat. sol. (a) 201:R49 (2004).

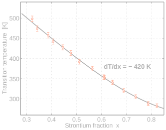
SBN – $Sr_xBa_{1-x}Nb_2O_6$ – Transition Temperature







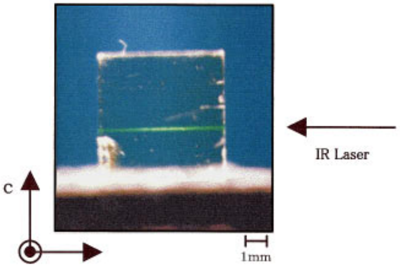




Ä. Andresen, A.-N. Bahar, D. Conradi, I.-I. Oprea, R. Pankrath, U. Voelker, K. Betzler, M. Wöhlecke, U. Caldiño, E. Martín, D. Jaque, J. García Solé: *Spectroscopy of Eu^{3+} ions in congruent strontium barium niobate crystals*. Phys. Rev. B 77:214102 (2008).

k-Space Spectroscopy

k-Space Spectroscopy – the Trigger



S. Kawai, T. Ogawa, H. S. Lee, Robert C. DeMattei, and Robert S. Feigelson:

Second-harmonic generation from needlelike ferroelectric domains in $Sr_{0.6}Ba_{0.4}Nb_2O_6$ single crystals.

Appl. Phys. Letters 73:768 (1998).

Explanation: Second-Harmonic Generation

$$\begin{pmatrix} E_1^{2\omega} \\ E_2^{2\omega} \\ E_3^{2\omega} \end{pmatrix} \propto \begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_1^{\omega} E_1^{\omega} \\ E_2^{\omega} E_2^{\omega} \\ E_3^{\omega} E_3^{\omega} \\ 2 E_2^{\omega} E_3^{\omega} \\ 2 E_3^{\omega} E_1^{\omega} \\ 2 E_1^{\omega} E_2^{\omega} \end{pmatrix}$$

Second-Harmonic Generation – *d*-Tensor

$$\begin{pmatrix} E_1^{2\omega} \\ E_2^{2\omega} \\ E_3^{2\omega} \end{pmatrix} \propto \begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_1^{\omega} E_1^{\omega} \\ E_2^{\omega} E_2^{\omega} \\ E_3^{\omega} E_3^{\omega} \\ 2E_2^{\omega} E_3^{\omega} \\ 2E_3^{\omega} E_1^{\omega} \\ 2E_1^{\omega} E_2^{\omega} \end{pmatrix}$$

$$d_{ik} = f(P)$$

Second-Harmonic Generation – *d*-Tensor

$$\begin{pmatrix} E_1^{2\omega} \\ E_2^{2\omega} \\ E_3^{2\omega} \end{pmatrix} \propto \begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_1^\omega E_1^\omega \\ E_2^\omega E_2^\omega \\ E_3^\omega E_3^\omega \\ 2E_2^\omega E_3^\omega \\ 2E_3^\omega E_1^\omega \\ 2E_1^\omega E_2^\omega \end{pmatrix}$$

$$d_{ik} = f(P) \stackrel{\text{try}}{=} a_0 + a_1 P + a_2 P^2 + a_3 P^3 + \dots$$

Second-Harmonic Generation – d-Tensor

$$\begin{pmatrix} E_1^{2\omega} \\ E_2^{2\omega} \\ E_3^{2\omega} \end{pmatrix} \propto \begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_1^{\omega} E_1^{\omega} \\ E_2^{\omega} E_2^{\omega} \\ E_3^{\omega} E_3^{\omega} \\ 2E_2^{\omega} E_3^{\omega} \\ 2E_3^{\omega} E_1^{\omega} \\ 2E_1^{\omega} E_2^{\omega} \end{pmatrix}$$

$$d_{ik} = f(P) \overset{\text{try}}{=} a_0 + a_1 P + a_2 P^2 + a_3 P^3 + \ldots, \quad a_0, a_2, \ldots = 0$$

Second-Harmonic Generation – *d*-Tensor

$$\begin{pmatrix} E_1^{2\omega} \\ E_2^{2\omega} \\ E_3^{2\omega} \end{pmatrix} \propto \begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_1^{\omega} E_1^{\omega} \\ E_2^{\omega} E_2^{\omega} \\ E_3^{\omega} E_3^{\omega} \\ 2E_2^{\omega} E_3^{\omega} \\ 2E_3^{\omega} E_1^{\omega} \\ 2E_1^{\omega} E_2^{\omega} \end{pmatrix}$$

$$d_{ik} = f(P) \overset{\text{try}}{=} a_0 + a_1 P + a_2 P^2 + a_3 P^3 + \ldots , \quad a_0, a_2, \ldots = 0$$

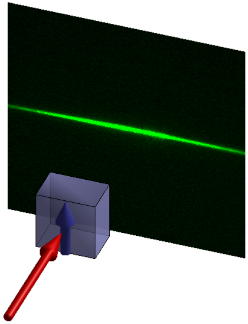
Second-Harmonic Generation as Polarization Probe

$$\begin{pmatrix} E_1^{2\omega} \\ E_2^{2\omega} \\ E_3^{2\omega} \end{pmatrix} \propto \begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_1^\omega E_1^\omega \\ E_2^\omega E_2^\omega \\ E_3^\omega E_3^\omega \\ 2E_2^\omega E_3^\omega \\ 2E_3^\omega E_1^\omega \\ 2E_1^\omega E_2^\omega \end{pmatrix}$$

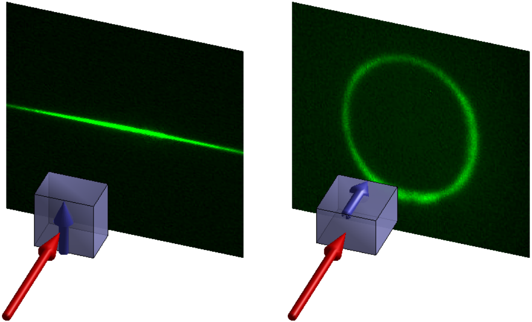
$$d_{ik} = f(P) \stackrel{\text{try}}{=} a_0 + a_1 P + a_2 P^2 + a_3 P^3 + \dots, \quad a_0, a_2, \dots = 0$$

$$E^{2\omega}(T) \Longrightarrow P(T)$$

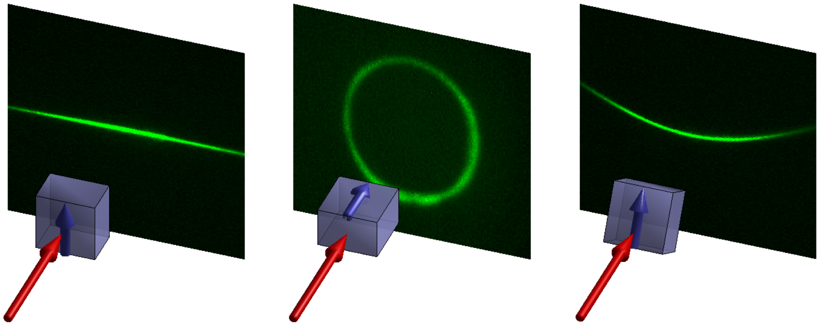
Geometrical Implications



Geometrical Implications

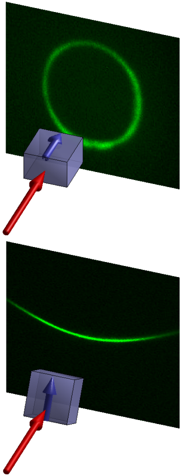


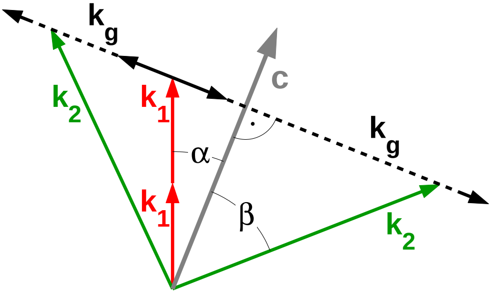
Geometrical Implications



Arthur R. Tunyagi, Michael Ulex, and Klaus Betzler: *Noncollinear optical frequency doubling in strontium barium niobate*, Physical Review Letters 90:243901 (2003).

Noncollinear Random Quasi Phase Matching



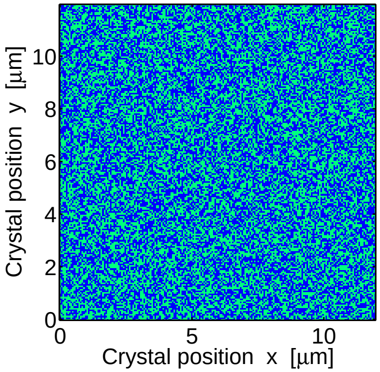


$$2\mathbf{k_1} + \mathbf{k_g} = \mathbf{k_2}$$

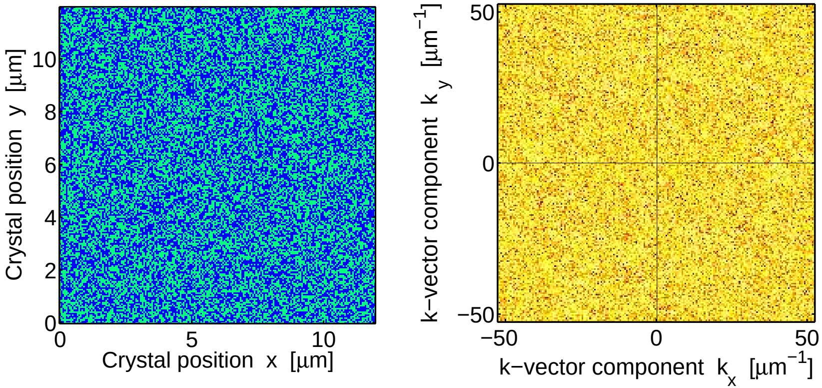
$$2|\mathbf{k_1}|\cos\alpha = |\mathbf{k_2}|\cos\beta$$

$$n_1(\alpha)\cos\alpha = n_2(\beta)\cos\beta \quad \text{(cone!)}$$

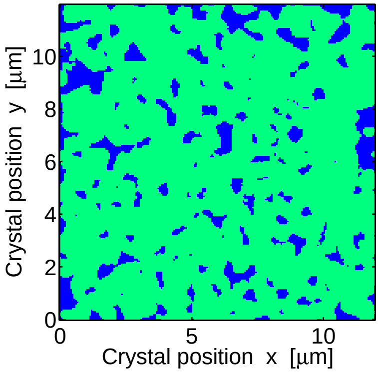
Real Space – Small Domains

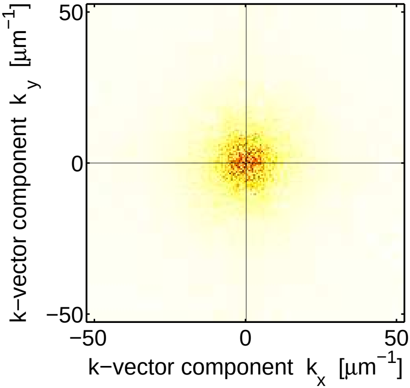


Real Space – Small Domains $\Longrightarrow$ k-Space

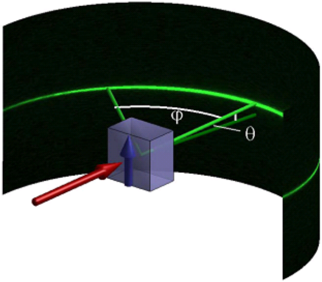


Real Space – Large Domains ⟹ k-Space

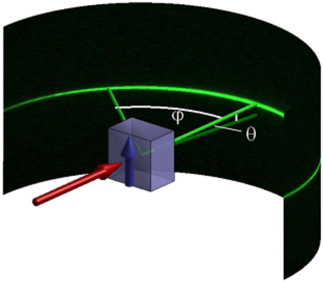




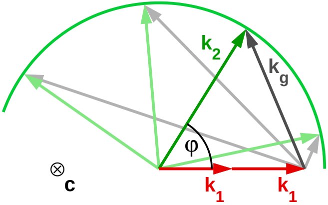
Accessible k-Spectrum



Accessible k-Spectrum

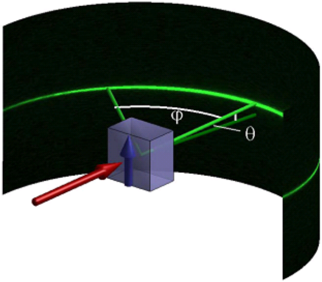


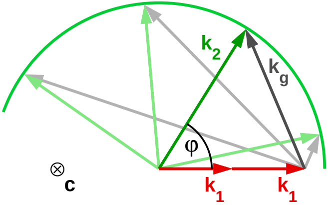
$$|\mathbf{k_g}| = \left(4|\mathbf{k_1}|^2 + |\mathbf{k_2}|^2 - 4|\mathbf{k_1}||\mathbf{k_2}|\cos\varphi\right)^{\frac{1}{2}}$$

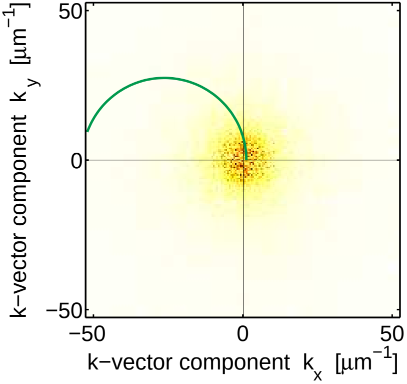


Accessible k-Spectrum

$$|\mathbf{k_g}| = \left(4|\mathbf{k_1}|^2 + |\mathbf{k_2}|^2 - 4|\mathbf{k_1}||\mathbf{k_2}|\cos\varphi\right)^{\frac{1}{2}}$$

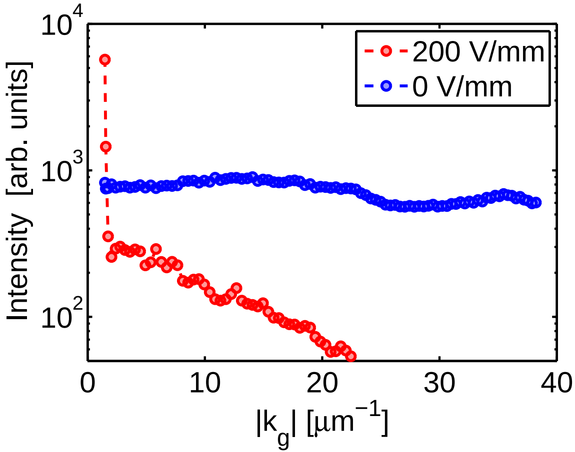






Results

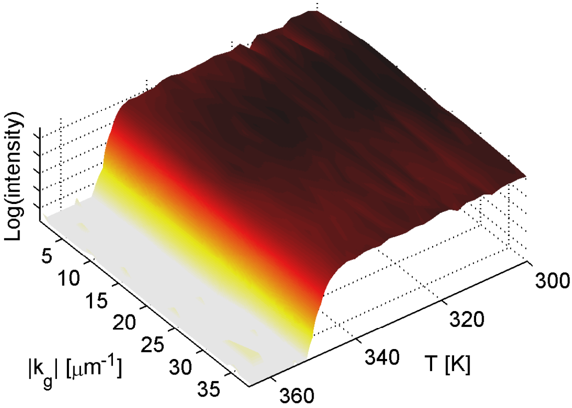
Poled and Unpoled States at Room Temperature



Uwe Voelker and Klaus Betzler: *Domain morphology from k-space spectroscopy of ferroelectric crystals.* Phys. Rev. B 74:132104 (2006).

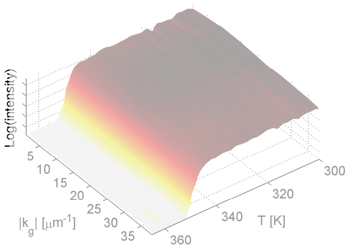
Temperature Dependence: Unpoled Sample

Heating an unpoled SBN sample



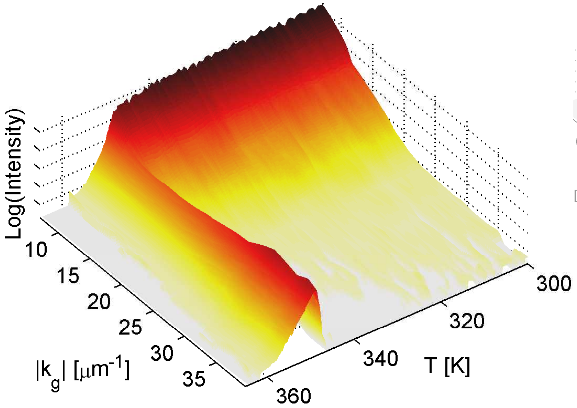
Temperature Dependence: Unpoled Sample

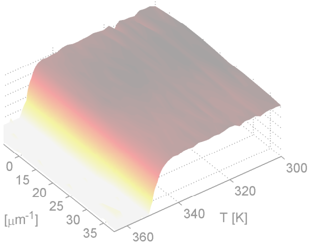
Heating an unpoled SBN sample



Temperature Dependence: Poled Sample

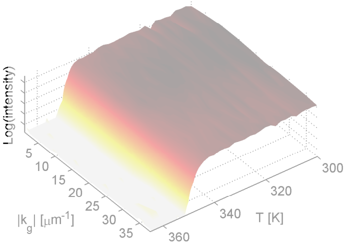
Heating a poled SBN sample





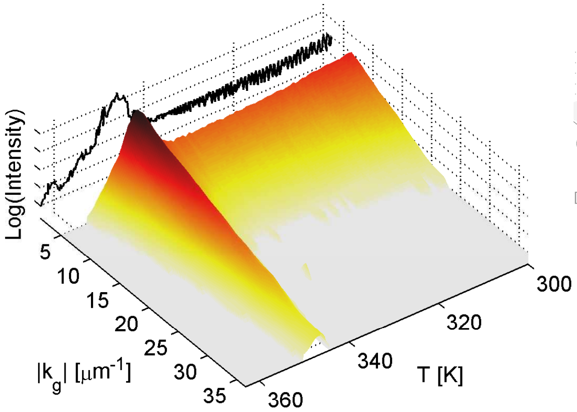
Temperature Dependence: Unpoled Sample

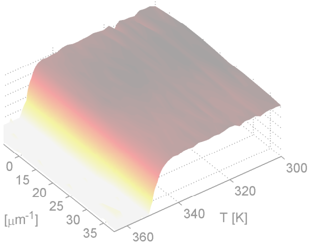
Heating an unpoled SBN sample



Temperature Dependence: Poled Sample

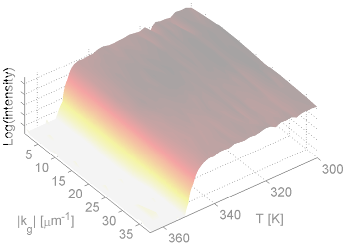
Heating a poled SBN sample (higher poling field)





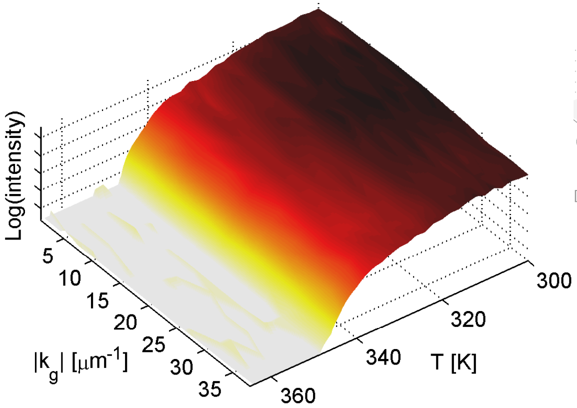
Temperature Dependence: Unpoled Sample

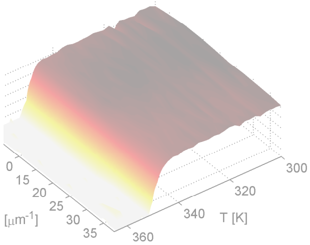
Heating an unpoled SBN sample



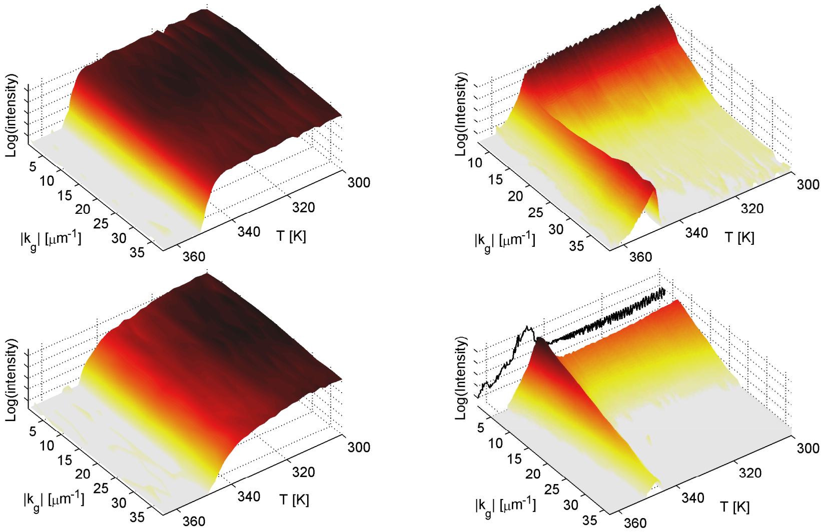
Temperature Dependence: Unpoled Sample

Cooling an unpoled SBN sample



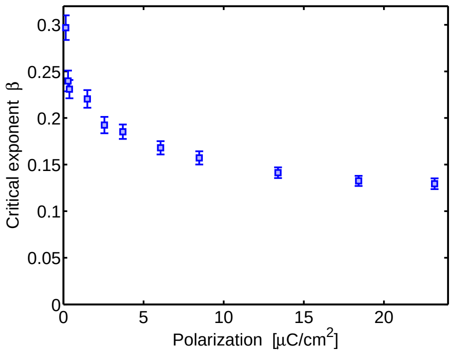


k-Space Fingerprints



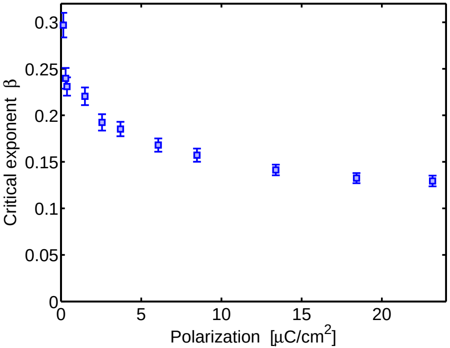
Preparation Dependence of the Phase Transition

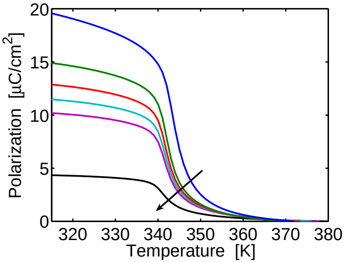
T. Granzow, Th.Woike, M.Wöhlecke, M. Imlau, W. Kleemann: *Change from 3D-Ising to Random Field-Ising-Model Criticality in a Uniaxial Relaxor Ferroelectric*. Phys. Rev. Letters 92:065701 (2004).



Preparation Dependence of the Phase Transition

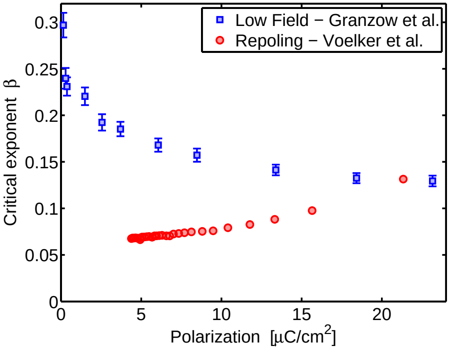
T. Granzow, Th.Woike, M.Wöhlecke, M. Imlau, W. Kleemann: *Change from 3D-Ising to Random Field-Ising-Model Criticality in a Uniaxial Relaxor Ferroelectric*. Phys. Rev. Letters 92:065701 (2004).

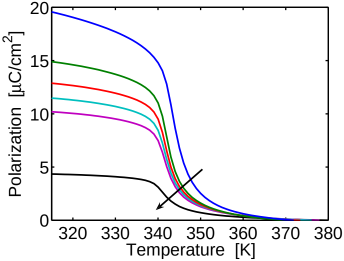




Preparation Dependence of the Phase Transition

T. Granzow, Th.Woike, M.Wöhlecke, M. Imlau, W. Kleemann: *Change from 3D-Ising to Random Field-Ising-Model Criticality in a Uniaxial Relaxor Ferroelectric*. Phys. Rev. Letters 92:065701 (2004).





Uwe Voelker, Urs Heine, Christoph Gödecker, Klaus Betzler: *Domain size effects in a uniaxial ferro-electric relaxor system: The case of $Sr_xBa_{1-x}Nb_2O_6$*. J. Appl. Phys. 102:114112 (2007).

Conclusions

Conclusions

- Results depend on . . .

Conclusions

- Results depend on . . .
- . . . sample preparation

Conclusions

- Results depend on . . .
- . . . sample preparation
- . . . sample history

Conclusions

- Results depend on ...
- ...sample preparation
- ...sample history
- ...type of measurement

Conclusions

- Results depend on . . .
- . . . sample preparation
- . . . sample history
- . . . type of measurement
- . . . velocity of measurement

Conclusions

- Results depend on . . .
- . . . sample preparation
- . . . sample history
- . . . type of measurement
- . . . velocity of measurement
- . . . polarization direction

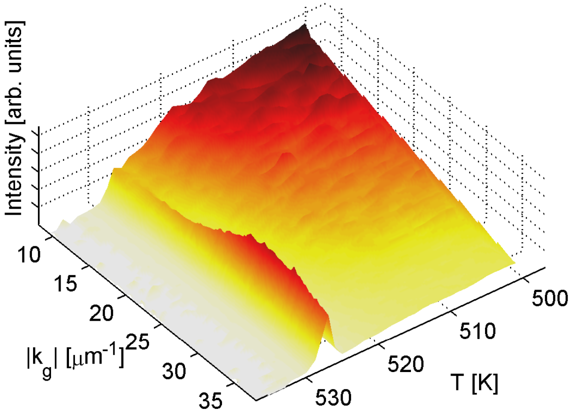
Conclusions

- Results depend on . . .
- . . . sample preparation
- . . . sample history
- . . . type of measurement
- . . . velocity of measurement
- . . . polarization direction
- . . . individual crystal ?

Special Case of SBN ?

Similar Results for Other Relaxors

Calcium barium niobate (CBN) – heating characteristics of a poled sample



Conclusions cont'd

- No unique phase transition of poled crystals

Conclusions cont'd

- No unique phase transition of poled crystals
- No thermodynamic equilibrium

Conclusions cont'd

- No unique phase transition of poled crystals
- No thermodynamic equilibrium
- No unique polarization in unpoled or partially-poled crystals

Conclusions cont'd

- No unique phase transition of poled crystals
- No thermodynamic equilibrium
- No unique polarization in unpoled or partially-poled crystals
- Any scaling attempts must fail

Conclusions cont'd

- No unique phase transition of poled crystals
- No thermodynamic equilibrium
- No unique polarization in unpoled or partially-poled crystals
- Any scaling attempts must fail
- Implications for critical exponents

Conclusions cont'd

- No unique phase transition of poled crystals
- No thermodynamic equilibrium
- No unique polarization in unpoled or partially-poled crystals
- Any scaling attempts must fail
- Implications for critical exponents
- Polarization directions locally not equivalent

Conclusions cont'd

- No unique phase transition of poled crystals
- No thermodynamic equilibrium
- No unique polarization in unpoled or partially-poled crystals
- Any scaling attempts must fail
- Implications for critical exponents
- Polarization directions locally not equivalent
- Global polarization no suitable order parameter ?

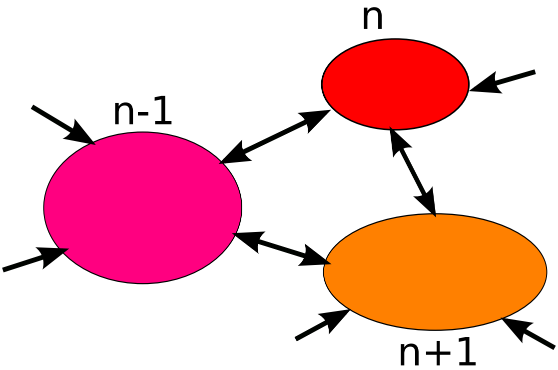
Conclusions cont'd

- No unique phase transition of poled crystals
- No thermodynamic equilibrium
- No unique polarization in unpoled or partially-poled crystals
- Any scaling attempts must fail
- Implications for critical exponents
- Polarization directions locally not equivalent
- Global polarization no suitable order parameter ?
- Free energy depending not only on *unique* P ?

Loosely Coupled Regions ?

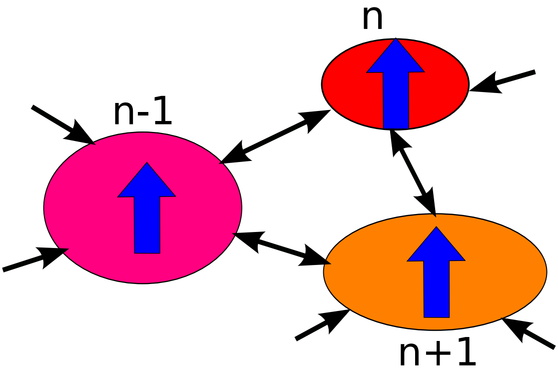
Loosely Coupled Regions ?

Varying composition, different structural stability



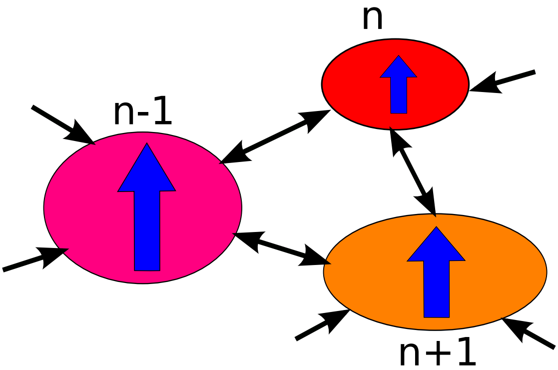
Loosely Coupled Regions ?

Unique polarization – rather unlikely



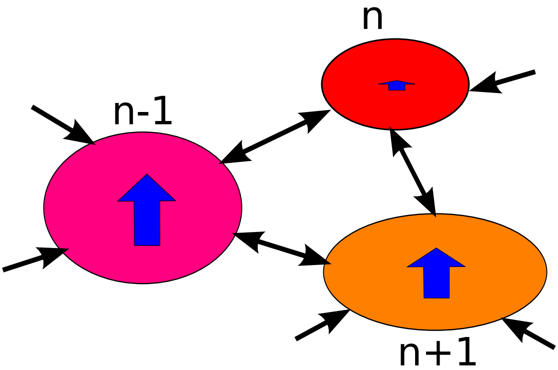
Loosely Coupled Regions ?

Different local polarization



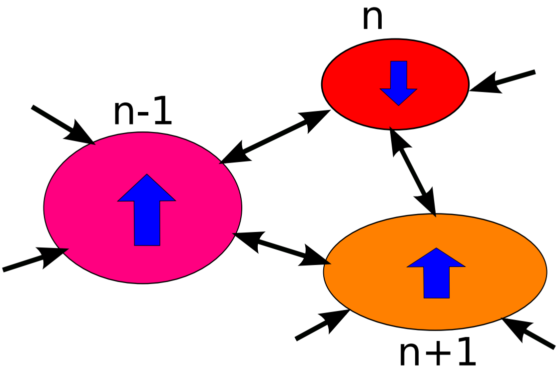
Loosely Coupled Regions ?

Near the phase transition



Loosely Coupled Regions ?

Polarization might be even locally reversed



Polarization as Order Parameter ?

Polarization as Order Parameter ?

no unique **P**(T) throughout the crystal

Polarization as Order Parameter ?

no unique $\mathbf{P}(T)$ throughout the crystal

$$\Downarrow$$

instead local $\mathbf{P}_n(T) \quad \Rightarrow \quad \mathbf{P}(T) = \int \mathbf{P}_n(T)\, dV$

Polarization as Order Parameter ?

no unique $\mathbf{P}(T)$ throughout the crystal

$$\Downarrow$$

instead local $\mathbf{P}_n(T) \quad \Rightarrow \quad \mathbf{P}(T) = \int \mathbf{P}_n(T)\, dV$

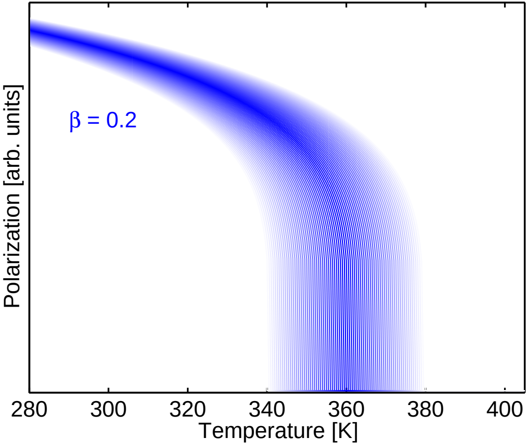
Additional terms in Hamiltonian due to

- Composition Variation
- Nonuniform Stress
- Nonequivalent Polarization Directions
- ...

Locally Different Transition Temperatures

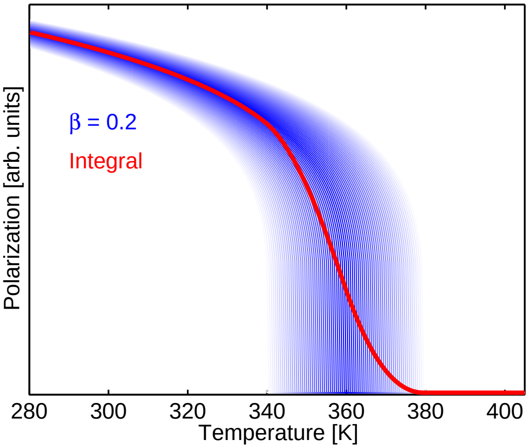
Locally Different Transition Temperatures

Polarization described by a unique critical exponent β



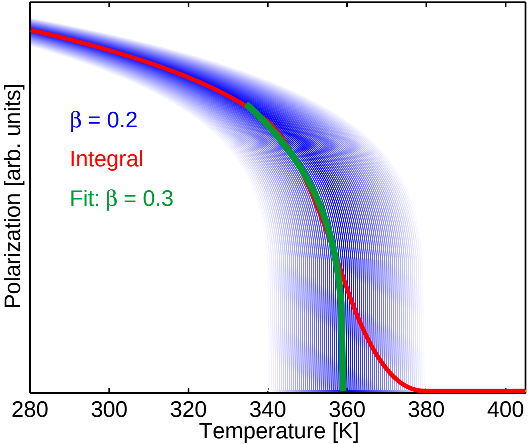
Locally Different Transition Temperatures

Global polarization as integral over the crystal



Locally Different Transition Temperatures

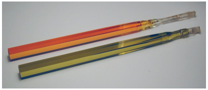
Critical exponent β pretended by an *excellent* fit



Thanks . . .

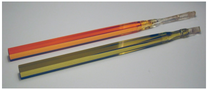
Thanks . . .

**to the crystal growers — Rainer Pankrath, Sergey Podlozhenov, Michael Ulex (SBN)
Manfred Mühlberg, Manfred Burianek (CBN)**



Thanks . . .

to the crystal growers — Rainer Pankrath, Sergey Podlozhenov, Michael Ulex (SBN) Manfred Mühlberg, Manfred Burianek (CBN)

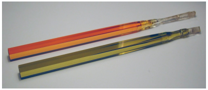


for financial support

Deutsche Forschungsgemeinschaft DFG

Thanks . . .

to the crystal growers — Rainer Pankrath, Sergey Podlozhenov, Michael Ulex (SBN) Manfred Mühlberg, Manfred Burianek (CBN)



for financial support

Deutsche Forschungsgemeinschaft DFG

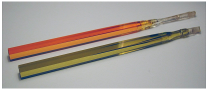
for the invitation

Penn University of Pennsylvania

ONR Office of Naval Research

Thanks . . .

to the crystal growers — Rainer Pankrath, Sergey Podlozhenov, Michael Ulex (SBN) Manfred Mühlberg, Manfred Burianek (CBN)



for financial support

Deutsche Forschungsgemeinschaft DFG

for the invitation

Penn University of Pennsylvania

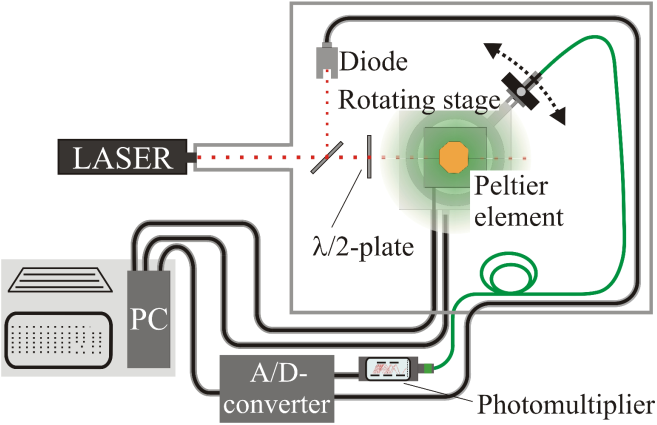
ONR Office of Naval Research

Thank you for your attention

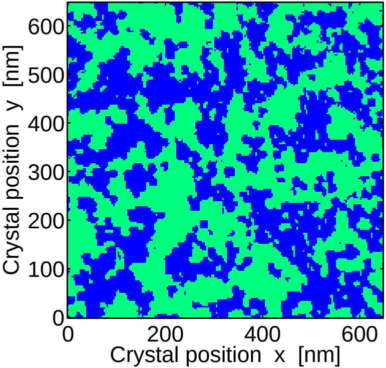
Additional Material

- Setup for k-Space Spectroscopy
- Calculated k-Space Representation of Real Domains
- Domain Lengths – Model Calculations
- Domain Lengths – Measurements
- k-Space Spectrum and Electric Field
- Conical Light Scattering at Higher Temperatures
- Beam Shape and its Fourier Transform

Setup for k-Space Spectroscopy

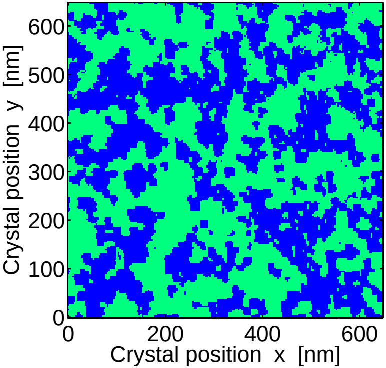


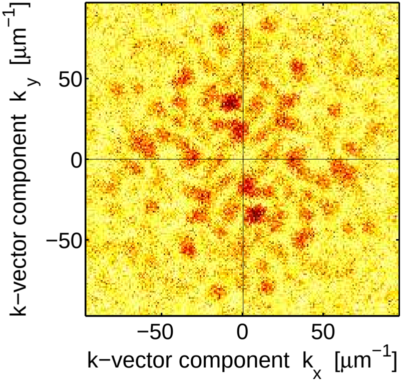
Calculated k-Space Representation of Real Domains



Real-space distribution taken from: P. Lehnen, W. Kleemann, Th. Woike, R. Pankrath:
Ferroelectric nanodomains in the uniaxial relaxor system $Sr_{0.61-x}Ba_{0.39}Nb_2O_6{:}Ce_x^{3+}$.
Physical Review B 64:224109 (2001).

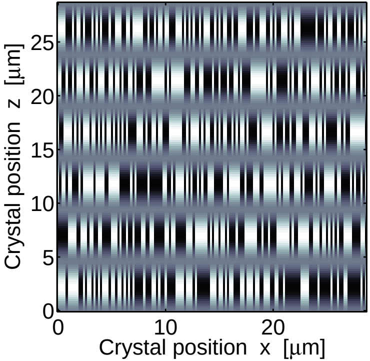
Calculated k-Space Representation of Real Domains



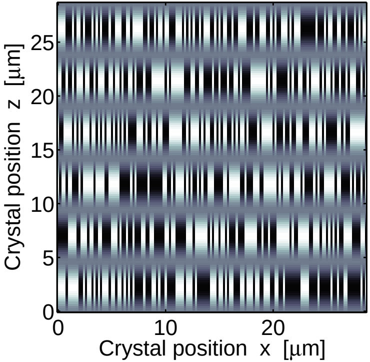


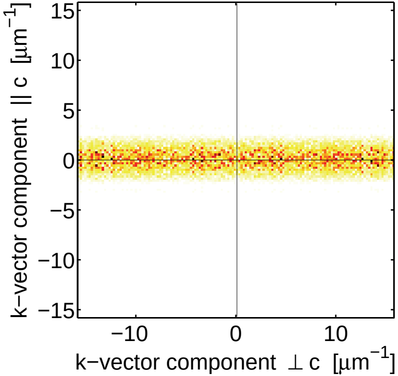
Real-space distribution taken from: P. Lehnen, W. Kleemann, Th. Woike, R. Pankrath: *Ferroelectric nanodomains in the uniaxial relaxor system* $Sr_{0.61-x}Ba_{0.39}Nb_2O_6{:}Ce_x^{3+}$. Physical Review B 64:224109 (2001).

Domain Lengths – Model Calculations



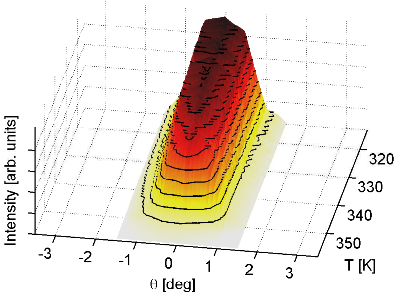
Domain Lengths – Model Calculations





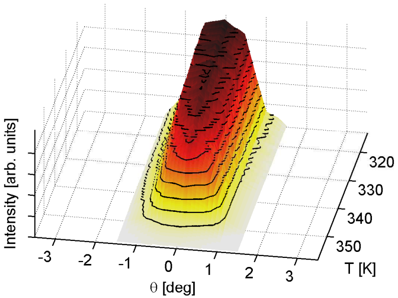
Domain Lengths – Measurement

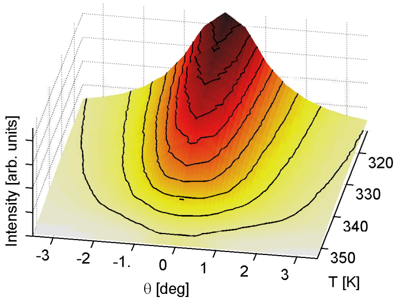
Poled sample – heating



Domain Lengths – Measurement

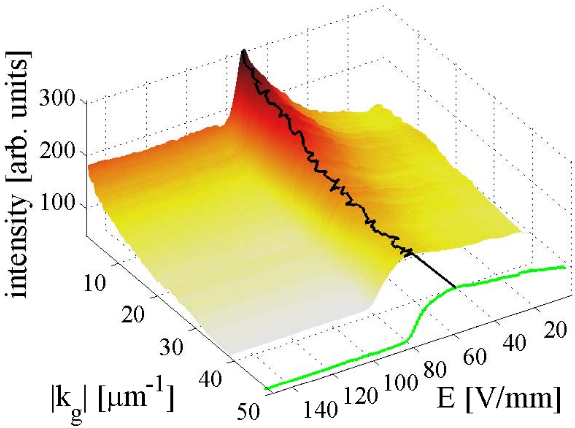
Poled sample – heating (left) and cooling (right)



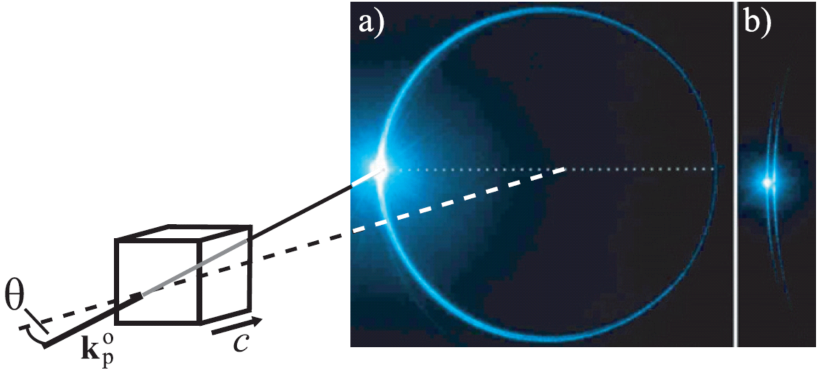


k-Space Spectrum and Electric Field

Application of an electric field to previously unpoled SBN

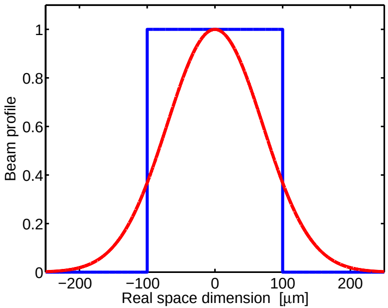


Conical Light Scattering at Higher Temperatures



K. Bastwöste, U. Sander, M. Imlau: *Conical light scattering in strontium barium niobate crystals related to an intrinsic composition inhomogeneity*. J. Phys.: Condens. Matter 19:156225 (2007).

Beam Shape and its Fourier Transform



Beam Shape and its Fourier Transform