Temperature Dependence of Short-Range Order in β-Brass

O. W. DIETRICH AND J. ALS-NIELSEN
The Danish Atomic Energy Commission, Research Establishment Risö, Roskilde, Denmark
(Received 31 May 1966; revised manuscript received 24 June 1966)

Critical scattering of neutrons around the superlattice reflections (1, 0, 0) and (1, 1, 1) from a single crystal of β-brass has been measured at temperatures from 2 to 25°C above the transition temperature. The temperature dependence of the critical peak intensity, proportional to the susceptibility χ, and that of the inverse correlation range κ_1 were, respectively, fitted to the power laws $\chi\propto((T-T_c)/T)^{-\gamma}$ and $\kappa_1=(k/a)\times((T-T_c)/T_c)^\nu$, yielding $\gamma=1.25\pm0.02$, $\nu=0.65\pm0.02$, and $k=3.0\pm0.3$. Here a is the cube edge of the unit cell. These results agree with the theoretical predictions of the Ising model.

INTRODUCTION

In the preceding paper[1] it was shown in detail that the data from critical scattering from β-brass at a fixed temperature above the transition temperature T_c were completely consistent with the classical Ornstein-Zernike correlation function proportional to $r_1^{-2}e^{-\kappa_1 r}/r$, which describes the correlation of occupation of lattice sites above T_c.

This paper deals with the temperature dependence of the inverse correlation length κ_1 and of the susceptibility analog χ proportional to $(r_1\kappa_1)^{-2}$. Susceptibility analog is here used to indicate that the order-disorder transition for occupation of lattice sites in an alloy is formally equivalent to the magnetic order-disorder transition in an Ising antiferromagnet. In recent years different theoretical approximate methods have revealed that

$$\chi\propto\left(\frac{T-T_c}{T}\right)^{-\gamma},\quad \frac{T-T_c}{T}\ll 1 \tag{1}$$

with[2] $\gamma=5/4$, in contrast to the result $\gamma=1$ obtained from more simple, classical theories (molecular-field theory, cluster theory[3-5]). The present experiment shows that $\gamma=1.25$ within approximately 2% accuracy.

The theoretical temperature dependence of κ_1 or of the dimensionless quantity $a\kappa_1$ (a being the cube edge of the unit cell) is also expressed by a power law[6]:

$$(a\kappa_1)=k\left(\frac{T-T_c}{T_c}\right)^{\nu},\quad \frac{T-T_c}{T_c}\ll 1, \tag{2}$$

and the classical theories give $\nu=\gamma/2=\frac{1}{2}$, whereas very recent calculations[7] by Fisher and Burford give $\nu=9/14$ from the relation $(2-\eta)\nu=\gamma$ with $\eta=1/18$ and $\gamma=5/4$. The parameter η describes, as mentioned in Ref. 1, a modified form of the Ornstein-Zernike pair correlation function

$$p(\mathbf{r})\propto e^{-\kappa_1 r}/r^{1+\eta}. \tag{3}$$

The experimental result on ν is $\nu=0.65\pm0.02$ with $\eta=1/18$. Also the theoretical value of k in (2) is compared with experiment.

EXPERIMENTAL PROCEDURE

Temperature Control

The crystal was mounted in an oven with a diameter large enough to prevent neutrons scattered from irradiated parts of the walls or the heating coil from hitting the BF_3 detector in a single scattering process. The oven was filled with He to obtain minimal temperature gradients, which were further reduced to less than 0.1°C by adjusting the currents in the three coils composing the heater. Furthermore, the He gave appropriate time constants for the electronic temperature control, which in its final version kept the temperature constant within ±0.05°C.

The crystal was tightly encapsuled in a thin stainless-steel container to avoid Zn evaporation. A 5% loss in Zn content implies a drop of 14°C of T_c, but in the duration of several months of the experiment, T_c remained constant at 466°C within 0.1°C. The temperature was measured by three Pt-(Pt-Rh) thermocouples placed on the outside of the container, at the top, the middle, and the bottom.

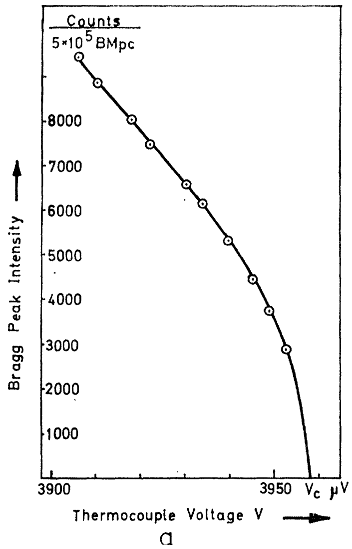
The critical temperature T_c was obtained by measuring the intensity I_B of the superlattice Bragg reflection for temperatures below T_c; see Fig. 1(a). The Bragg intensity is proportional to the square of the long-range-order parameter which vanishes at T_c, and the intersection with the temperature axis will therefore give the thermocouple voltage V_c when the crystal is in the critical state. The intersection V_c was determined by a least-squares fit of I_B to a power law

$$I_B=c(V_c-V)^\beta$$

by requiring a minimal value of

$$S^2\equiv\textstyle\sum_i w_i\{I_i-c(V_c-V)^\beta\}^2,$$

[1] J. Als-Nielsen and O. W. Dietrich, preceding paper, Phys. Rev. **153**, 706 (1966).
[2] C. Domb and M. F. Sykes, Proc. Roy. Soc. (London) **A240**, 214 (1957); J. Math. Phys. **2**, 63 (1961); M. F. Sykes, *ibid.* **2**, 52 (1961); G. A. Baker, Phys. Rev. **124**, 768 (1961).
[3] R. J. Elliott and W. Marshall, Rev. Mod. Phys. **30**, 75 (1958).
[4] J. M. Cowley, Phys. Rev. **120**, 1648 (1961).
[5] F. Zernike, Physica **7**, 565 (1940).
[6] M. Fisher, in *Proceedings of the International Conference on Magnetism, Nottingham, 1964* (The Institute of Physics and The Physical Society, London, 1965), p. 81. M. Fisher, in Proceedings of the Conference on Phenomena in the Neighborhood of Critical Points, National Bureau of Standards, Washington, D. C., 1965 (unpublished).
[7] M. Fisher (private communication).



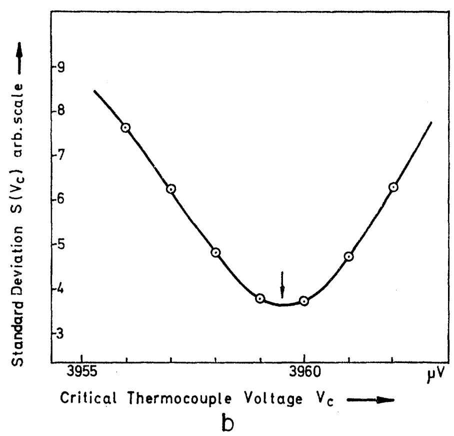


FIG. 1. (a) Shows the Bragg peak intensity I_B at temperatures below T_c. The full line is a weighted least-squares fit of I_B to the power law $I_B \propto (V_c - V)^\beta$. (b) Shows the standard deviation of the fit for different choices of V_c. The arrow thus indicates the critical thermocouple voltage V_c. The uncertainty of V_c is estimated to be approximately 1 μV ≈ 0.1°K. (BMpc = beam-monitor preset count.)

w_i denoting the statistical weight on the measured value I_i. $S(V_c)$ is shown in Fig. 1(b). It is concluded that the uncertainty of V_c is approximately 1 μV. The temperature dependence of all parameters to be discussed is expressed by the temperature difference $\Delta T = T - T_c$, and this is measured as the corresponding difference in thermocouple voltage multiplied by the scaling factor 0.103°K/μV. The uncertainty of V_c contributes the main part of the uncertainty ±0.1° on ΔT.

Samples

Three different single crystals of approximately 1-in. linear dimensions were examined, but analysis of the composition showed that only one of them (No. III) was a pure β-brass single crystal. The major part of the results discussed here derive therefore from that crystal. Results from crystal No. I have been reported on earlier[8] and are only briefly recapitulated in the Appendix. This crystal contained a considerable amount of γ phase which was apparent from metallurgical examination, chemical analysis, and perhaps most clearly from a neutron diffraction pattern measured along the (1,0,0) direction; see Fig. 2. The composition of crystal No. II was similar to that of No. I, and the results from these two crystals are in agreement, as seen in the Appendix.[9]

RESULTS

Inverse Correlation Range $\kappa_1(\Delta T/T_c)$

The inverse correlation range κ_1 can in principle be deduced from any scan through a superlattice reflection above T_c. The geometry of our setup favored, however, the type of scan in which the crystal is rotated around a vertical axis (y scan in the notation of Ref. 1). For a given wavelength the resolution correction is smallest for the (1,0,0) reflection, since the angular widths of the resolution curves have to be multiplied by $|\boldsymbol{\tau}|$ for conversion to Å^{-1}. Therefore y scans at 2.70 Å through the (1,0,0) reflection will give the most accurate determination of κ_1. Results from the pure β-brass crystal are shown in Fig. 3. The results for all three crystals are tabulated in the Appendix.

Check Possibilities

(*1*) *Different types of scan.* In the Appendix are also listed the results of 1:2 scans through (1,0,0), (x scans). Agreement with y scans was found.

(*2*) *Different reflections.* Also y scans through the (1,1,1) reflection were measured and the results agree with (*1*); (*1*) and (*2*) give support to the assumption that the cross section is isotropic around each superlattice reflection.

(*3*) *Different wavelengths.* As mentioned in Ref. 1, a wavelength of 1.35 Å could be obtained without changing the geometry. Again, satisfactory agreement was obtained at several temperatures, and we conclude therefore that the static approximation is valid and that the constant-background-subtraction procedure, which might be erroneous due to phonon contributions, is sufficiently correct. This was also proved by a scan taken 70°C above T_c, because the intensity when corrected for the small amount of remaining critical scattering appeared to be constant.

In addition to the conclusions drawn from (*1*), (*2*), and (*3*) it can be generally concluded that the resolution correction must be correct, since it varies in the above-mentioned cases. However, this conclusion was checked with (*4*) below.

---

[8] O. W. Dietrich and J. Als-Nielsen, in Proceedings of the Conference on Phenomena in the Neighborhood of Critical Points, National Bureau of Standards, Washington, D. C., 1965 (unpublished).

[9] We appreciate the loan of specimens II and III from Chalk River Nuclear Laboratories, where the phonon spectrum for these specimens had just been examined. See G. Gilat and G. Dobling, Phys. Rev. **138**, A1053 (1965).

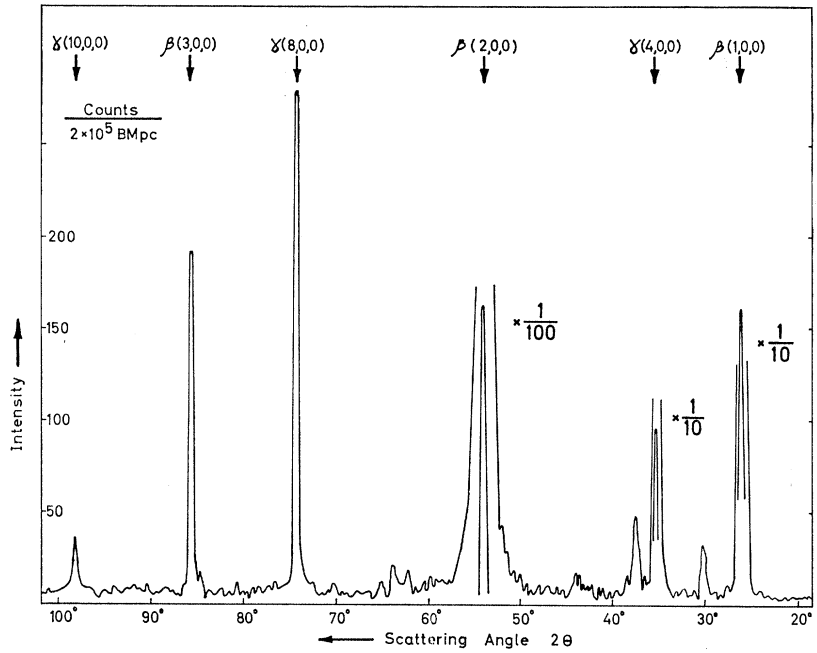


FIG. 2. Neutron diffraction pattern at 440°C along the (1,0,0) direction in reciprocal space for specimen II. The content of γ phase in the crystal is clearly demonstrated. The diffraction pattern for specimen I was similar to that in Fig. 2, whereas the γ peaks were absent in the diffraction pattern for specimen III.

(*4*) *Different collimation.* The most pronounced reduction in resolution correction was obtained by horizontal Soller collimation in the second collimator. The result of a scan with this collimation (Table III) agrees perfectly with the above-mentioned results, giving the final proof for the accuracy of the unfolding procedure described in Ref. 1.

Uncertainties

The uncertainty of the values of κ_1 can only be estimated. The statistical uncertainty on κ_1 from the least-squares fit was in most cases around 1% and did not in any case exceed 2%. The uncertainty in $\Delta T/T_c$ converted to uncertainty in κ_1 was estimated to be approximately 3% for $\Delta T/T_c = 3\times10^{-3}$ and around 0.3% for $\Delta T/T_c = 30\times10^{-3}$. Additional sources of uncertainty in κ_1 are the background subtraction and the resolution correction. It is concluded that the resulting uncertainties of the values of κ_1 are between 2% and 5%. The dashed line in Fig. 3 is a least-squares fit with equal weights on all points to the power law (2), resulting in

$$\nu = 0.647 \pm 0.022\,,$$
$$k = 2.99 \pm 0.32\,,$$

Note that all uncertainties are standard deviations.

TABLE I. Theoretical values for a bcc lattice of k and ν in $(a\kappa_1) = k(T-T_c/T_c)^\nu$, $T-T_c/T_c \ll 1$ a is the cube edge of the unit cell.

Theory	k	ν
Molecular-field theory	2.828	1/2
Cowley[a]	2.828	1/2
Zernike[b]	2.530	1/2
Elliott and Marshall[c]	2.432	1/2
Fisher and Burford[d]	2.565	9/14

[a] From A. Paskin, Phys. Rev. **134**, A246 (1964). See in particular footnote on page A248.
[b] From C. B. Walker and D. T. Keating, Phys. Rev. **130**, 1726 (1963), Fig. 6.
[c] Reference 3.
[d] Reference 7.

Comparison with Theory

Molecular-field theory, the cluster theory of Elliott and Marshall,[3] and similar theories of Cowley[4] and of Zernike[5] all give $\nu = \frac{1}{2}$, but different values of k in (2).

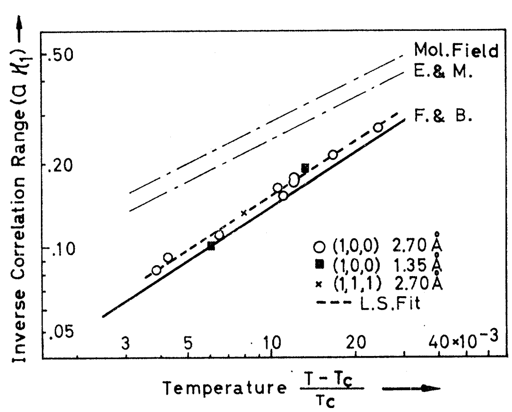


FIG. 3. Double log plot of $(a\kappa_1)$ versus $\Delta T/T_c$. The dashed line is a least-squares fit of the experimental points to the power law $(a\kappa_1) = k(\Delta T/T_c)^\nu$, giving $k = 2.99 \pm 0.32$ and $\nu = 0.65 \pm 0.02$. Also the theoretical curves of molecular-field theory, cluster theory of Elliott and Marshall, and lattice statistics theory of Fisher and Burford are shown. (L.S. = least squares.)

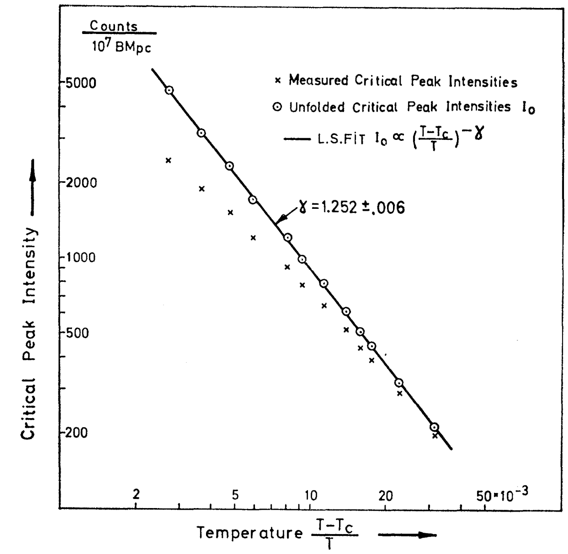


FIG. 4. Temperature dependence of peak intensity of critical scattering, proportional to the susceptibility χ of an Ising magnet. The theoretical power law $\chi \propto (\Delta T/T)^{-5/4}$ is verified within 2% accuracy, including estimated systematic errors.

For a bcc lattice their values of k are listed in Table I. It is apparent from Fig. 3 that the common temperature dependence of these theories is too crude. Fisher and Burford[6] have reported on calculations of $(a\kappa_1)$, based on series expansions in powers of $v = \tanh(J/kT)$ for temperatures down to $\Delta T/T_c = 0.03$ for the simple cubic lattice. Recently these authors have obtained an expression for $(a\kappa_1)$ for a bcc lattice, valid for temperatures much closer to T_c. Their result is given here[7]:

$$\ln(\kappa_1 a')^2 = \frac{9}{7}\ln\left(1-\frac{v}{v_c}\right) + \ln\frac{35}{48} - \ln v + \sum_{n=1}^{7} d_n\left(\frac{v}{v_c}\right)^n + \mathcal{R}_7\left(\frac{v}{v_c}\right); \quad (4)$$

a' is the distance between nearest neighbors,

$$v_c = \tanh(J/kT_c) = 0.156172\,,$$

$$d_1 = 0.03633,\ d_2 = 0.03311,\ d_3 = -0.00819,\ d_4 = 0.00705,$$
$$d_5 = -0.00313,\quad d_6 = 0.00345,\quad \text{and}\quad d_7 = -0.00169.$$

The remainder $\mathcal{R}_7(1)$ is estimated to be less than 0.0012. The derivation of this formula is based upon the pair-correlation function (3), which theoretically is believed to be a more accurate expression than the Ornstein-Zernike correlation function obtained with $\eta = 0$. In Ref. 1 it was concluded that it was not possible from the experimental data at a fixed temperature to determine whether $\eta = 1/18$ or $\eta = 0$. However, to compare the experimental absolute values of $(a\kappa_1)$ with (2), the data in Fig. 3 are analyzed with $\eta = 1/18$, as described in Ref. 1. Within the temperature region of the experimental results, (4) can, within an accuracy of 0.5%, be approximated with the simpler form (2), giving $\nu = 9/14 = 0.6429$ and $k = 2.565$. These theoretical values agree remarkably well with the experimental results. The difference of 17% in k between theory and experiment might be due to the only-nearest-neighbor interaction in the theoretical model, but it cannot be excluded that results from another β-brass crystal might differ some percent from the present results, because of different individual strains etc. in the samples.

Susceptibility-Analog $\chi(\Delta T/T)$

The susceptibility-analog χ is related to the pair-correlation function by

$$\chi \propto \sum_{\mathbf{r}} p(\mathbf{r})$$

in the notation adopted in Ref. 1. It is seen from (9) in Ref. 1 that the critical peak intensity I_0 (corrected for the resolution reduction) is also proportional to $\sum_{\mathbf{r}} p(\mathbf{r})$.

The exponent γ in (1) can therefore be determined from the measured temperature dependence of I_0, independently of any details in shape $p(\mathbf{r})$ may have, provided, of course, that the resolution correction is accurate. Our result for γ is therefore expected to remain valid even if more accurate experiments in the future should reveal a small deviation from the Ornstein-Zernike correlation function. γ could in principle be deduced from the critical scans, for example, around (1,0,0) at 2.70 Å, leading to the values of κ_1 in Fig. 3. However, these scans took several weeks of measuring time and possible long-term drift of electronics etc. might influence the accuracy of γ [but not so much $\kappa_1(\Delta T/T_c)$]. Therefore a separate peak-value run was undertaken to avoid this kind of error and furthermore to improve the statistics. The results are shown in Fig. 4 in a double log plot. The measured peak intensities were unfolded using κ_1 values from Table IV in the Appendix and were corrected for the slight change in Debye-Waller factor over the temperature region.

A least-squares fit gave $\gamma = 1.252 \pm 0.006$. This uncertainty expresses the scatter of measured points around the full line in Fig. 4, but it is not the uncertainty of γ, since it does not express a possible systematic error due to a slightly incorrect value of T_c, of the background, and of the peak-value reduction. Conservative estimates of these sources of error give the final result $\gamma = 1.25 \pm 0.02$ from the (1,0,0) peak-value run at 2.70 Å.

The reliability of this result is supported by similar results, summarized in Table IV, obtained at 1.35 Å and at the (1,1,1) reflection. Although η cannot be determined from the shape of a critical scattering curve, as mentioned in Ref. 1, our results on γ and ν nevertheless experimentally indicate a small positive value of η as predicted by theory, using the relation $(2-\eta)\nu = \gamma$:

$$\eta = 0.077 \pm 0.067\,.$$

The theoretical result on η is

$$\eta \approx 0.056.$$

CONCLUSION

The order-disorder transition in β-brass proved experimentally to be well described by the Ising model. Considerable theoretical efforts have in recent years been devoted to the development of an accurate statistical treatment of cooperative phenomena in the vicinity of the transition temperature using this model. The present experiment is significant in that some of the theoretical predictions have been verified for the first time.

Thus the temperature dependence of the peak intensity I_0 of critical scattering in a superlattice reflection fitted a power law $I_0 \propto (\Delta T/T)^{-1.25\pm0.02}$, verifying the 5/4 law for the susceptibility-analog. Also the inverse correlation range κ_1 fitted a power law $\kappa_1 \propto (\Delta T/T_c)^{+0.65\pm0.02}$, in agreement with the "9/14-law" of Fisher and Burford. Furthermore, the absolute values of κ_1 agree, within approximately 17%, with their predictions.

It is interesting to note that the exponents in the temperature dependences of I_0 and κ_1 give an experimental indication of a small positive value of η. Accepting the theoretical relation $(2-\eta)\nu=\gamma$, it is found that $\eta = 0.077\pm0.067$, which should be compared to the theoretical value of $\eta \approx 1/18$.

When the sample contains more Zn than permissible in the pure β phase, the experiment indicates that the exponents γ and ν are reduced significantly. However, a systematic investigation of this effect has not yet been undertaken. As far as we know, no theoretical prediction of this kind has been published as yet.

ACKNOWLEDGMENTS

We wish to express our appreciation for the encouragement of Professor O. Kofoed-Hansen throughout the work described in this and the preceding paper. We are indebted to Dr. Walter Marshall for discussions during the experiment and for originally directing our attention to this interesting field. Professor M. Fisher kindly discussed his calculation of $\kappa_1(T)$ in advance of publication. We appreciate the help of Dr. G. Dolling, who lent us the crystals. Our colleagues J. Linderholm and K. Christensen rendered valuable assistance throughout the experiment described in both these papers.

APPENDIX

Here we present tabulated experimental data on three β-brass single crystals (Tables II–IV). However, specimens I and II contained a few percent of γ phase.

The column headings are explained in detail as follows:

Critical Peak Intensities

λ is the monochromatic neutron wavelength used in the peak-value run. The background count rate B was approximately constant at all temperatures. $(\sigma_x,\sigma_y,\sigma_z)$ denote the resolution widths as measured by three perpendicular scans through the Bragg reflection below the critical temperature T_c. $\Delta T = T - T_c$ is the temperature deviation from the critical temperature. $\langle I \rangle$ is the

TABLE II. Experimental data for a 1-in.-diam spherical β-brass crystal with a few percent content of γ phase. $T_c = 468°C$.

	Critical peak intensities												
λ / Reflection / background / $(\sigma_x,\sigma_y,\sigma_z)$	2.70 Å / (1,0,0) / $331c/2\times10^7$ BMpc[a] / $(0.58, 0.70, 3.13)\times10^{-2}$ Å^{-1}				2.70 Å / (1,1,1) / $314c/2\times10^7$ BMpc[a] / $(0.47, 1.53, 3.03)\times10^{-2}$ Å^{-1}				Inverse correlation range $(a\kappa_1)$				
	$(\Delta T/T)$ $\times10^3$	$\langle I \rangle$	n	I_0	$(\Delta T/T)$ $\times10^3$	$\langle I \rangle$	n	I_0	$(\Delta T/T_c)$ $\times10^3$	Reflection	Type of scan	λ Å	$(a\kappa_1)$
	2.54	2898	6	4630(−1.6)	2.22	2357	9	4178(−5.3)	2.29	(1,0,0)	x	2.70	0.071(+8.9)
	4.02	2082	6	2670(−5.8)	3.61	1780	8	2428(−4.7)	2.50	···	···	···	0.085(−3.8)
	4.42	1919	5	2370(−7.2)	5.40	1463	10	1656(+2.9)	2.70	···	···	···	0.096(−11.5)
	7.22	1403	6	1410(−5.1)	8.00	1114	9	1038(+0.9)	3.91	···	···	···	0.108(−1.7)
	11.71	1079	5	900(+3.5)	11.96	882	5	678(+3.8)	4.25	(1,1,1)	y	···	0.114(−2.2)
	16.68	877	5	622(+5.6)	18.31	658	9	386(−4.2)	5.26	(1,0,0)	x	···	0.125(+1.1)
	22.24	716	5	424(−1.0)	28.79	542	8	245(+1.6)	5.40	···	···	···	0.134(−4.3)
	31.42	588	8	275(−6.1)					5.94	···	y	···	0.134(+1.2)
	46.46	501	14	179(−5.9)					7.69	(1,1,1)	y	···	0.148(+6.3)
									7.96	···	···	···	0.140(+13.9)
									10.66	···	···	···	0.197(−3.1)
									11.06	(1,0,0)	x	···	0.192(+1.6)
									14.84	···	···	···	0.238(−2.7)
									24.82	···	···	···	0.294(+6.2)
									35.61	···	···	···	0.425(−9.4)
									49.10	···	···	···	0.466(+0.1)
Results of weighted least-squares fits	$\gamma = 1.10\pm0.03$ including systematic errors				$\gamma = 1.13\pm0.02$ including systematic errors				$\nu = 0.58\pm0.02$				$k = 2.72\pm0.24$

[a] Beam-monitor preset count.

TABLE III. Experimental data for a $\frac{3}{4}$-in.-diam cylindrical β-brass crystal with a few percent content of γ phase (see Fig. 2). $T_c=468°C$.

	Critical peak intensities								Inverse correlation range $(a\kappa_1)$				
λ	2.70 Å				1.35 Å								
Reflection	(1,0,0)				(1,0,0)								
background	$324c/2\times10^7$ BMpc[a]				$693c/2\times10^7$ BMpc[a]								
$(\sigma_x,\sigma_y,\sigma_z)$	$(0.63, 0.89, 4.07)\times10^{-2}$ Å^{-1}				$(0.61, 0.89, 7.37)\times10^{-2}$ Å^{-1}								
	$(\Delta T/T)\times10^3$	$\langle I\rangle$	n	I_0	$(\Delta T/T)\times10^3$	$\langle I\rangle$	n	I_0	$(\Delta T/T_c)\times10^3$	Reflection	Type of scan	λ Å	$(a\kappa_1)$
	1.53	5049	8	12 501(−4.4)	2.22	4507	6	16 404(+3.9)	3.88	(1,0,0)	y	2.70	0.094(+5.3)
	1.74	4279	8	9907(−14.1)	4.30	3427	6	7790(+1.4)	4.45	···	y[b]	···	0.115(−7.4)
	3.05	3456	8	6100(+2.0)	7.08	2749	6	4515(+1.4)	5.26	···	y	···	0.125(−6.5)
	4.30	2610	7	3956(−2.7)	10.60	2251	6	2861(−1.3)	6.54	···	y	···	0.125(+5.5)
	7.53	1788	9	2129(−1.3)	14.27	1901	6	1980(−4.6)	6.60	···	y[b]	···	0.121(+9.2)
	12.10	1302	8	1275(+1.0)	19.65	1723	6	1522(+4.0)	7.99	···	y	···	0.150(−1.7)
	18.44	989	9	802(+2.3)					10.70	···	y	···	0.180(−3.8)
	31.02	717	9	443(+1.6)					12.95	···	y	1.35	0.196(−1.8)
	49.10	542	8	236(−0.9)					17.94	···	y	2.70	0.230(+0.2)
									23.06	···	y	···	0.261(+1.4)
Results of weighted least-squares fits	$\gamma=1.13\pm0.02$ including systematic errors				$\gamma=1.09\pm0.03$ including systematic errors				$\nu=0.55\pm0.03$				$k=2.12\pm0.30$

[a] Beam-monitor preset count.

[b] Horizontal collimation.

TABLE IV. Experimental data for a 1-in.-diam cylindrical pure β-brass crystal. $T_c=466°C$.

	Critical peak intensities																Inverse correlation range $(a\kappa_1)$					
λ	2.70 Å				2.70 Å				1.35 Å				1.35 Å									
Reflection	(1,0,0)				(1,1,1)				(1,0,0)				(1,1,1)									
background	$149c/10^7$ BMpc[a]				$177c/10^7$ BMpc[a]				$1164c/5\times10^7$ BMpc[a]				$857c/2\times10^7$ BMpc[a]									
$(\sigma_x,\sigma_y,\sigma_z)$	$(0.64, 0.48, 3.01)\times10^{-2}$ Å^{-1}				$(0.36, 0.66, 3.14)\times10^{-2}$ Å^{-1}				$(1.72, 0.39, 5.64)\times10^{-2}$ Å^{-1}				$(1.25, 0.72, 6.37)\times10^{-2}$ Å^{-1}								$(a\kappa_1)$	
	$(\Delta T/T)\times10^3$	$\langle I\rangle$	n	I_0	$(\Delta T/T)\times10^3$	$\langle I\rangle$	n	I_0	$(\Delta T/T)\times10^3$	$\langle I\rangle$	n	I_0	$(\Delta T/T)\times10^3$	$\langle I\rangle$	n	I_0	$(\Delta T/T_c)\times10^3$	Reflection	Type of scan	λ	$\eta=0$	$\eta=1/18$
	2.63	2614	40	4717(−0.6)	3.61	1244	6	1814(−7.1)	2.97	5515	6	13 378(+2.0)	3.13	3866	6	9340(−3.3)	3.86	(1,0,0)	y	2.70	0.0870(−1.3)	0.0835(−1.8)
	3.58	2041	6	3182(−1.8)	4.30	1087	6	1448(−7.5)	5.03	4263	6	7063(+0.4)	3.57	3622	6	7910(−3.6)	4.25	···	···	2.70	0.0962(−5.2)	0.0924(−5.6)
	4.65	1675	6	2341(−0.0)	5.64	921	6	1086(−2.0)	7.50	3493	5	4393(−0.3)	4.78	3165	6	5635(−1.6)	6.04	···	···	1.35	0.1051(+8.3)	0.1010(+8.2)
	5.79	1361	6	1733(−2.6)	6.66	829	6	906(+0.7)	10.05	3015	6	3096(−0.9)	6.09	2778	6	4139(−2.7)	6.51	···	···	2.70	0.1171(+2.3)	0.1111(+3.5)
	7.96	1071	6	1211(+1.4)	8.39	686	6	667(−1.0)	12.83	2689	6	2337(−0.3)	7.91	2479	6	3114(+1.1)	8.06	(1,1,1)	···	···	0.1375(−0.3)	0.1325(−0.3)
	9.17	931	6	993(−0.8)	9.74	632	6	577(+3.2)	14.89	2523	6	1982(+0.9)	9.49	2309	6	2583(+4.6)	10.55	(1,0,0)	···	···	0.1684(−3.4)	0.1631(−3.6)
	11.15	800	6	795(+1.4)	11.91	530	6	429(−1.0)					11.08	2096	6	2077(+2.0)	11.06	···	···	···	0.1586(+5.6)	0.1533(+5.7)
	13.75	673	12	617(+2.3)	14.25	490	6	369(+6.4)					13.85	1898	6	1615(+4.2)	12.04	···	···	···	0.1827(−3.2)	0.1767(−3.1)
	15.68	592	12	511(−0.1)	16.51	443	6	307(+6.4)					13.93	1845	17	1529(−0.4)	12.09	···	···	···	0.1758(+0.9)	0.1702(+0.9)
	17.46	544	12	449(+0.5)	18.42	428	6	285(+12.2)					17.73	1584	6	1044(−9.3)	13.31	···	···	1.35	0.1975(−4.6)	0.1916(−4.7)
	22.63	439	12	320(−1.0)	23.42	332	12	171(−8.0)					22.43	1519	6	891(+4.1)	16.72	···	···	2.70	0.2174(+0.3)	0.2115(+0.2)
	31.16	347	12	212(−1.8)	31.61	288	12	119(−6.1)					31.20	1306	10	561(−1.5)	24.42	···	···	2.70	0.2753(+0.7)	0.2694(+0.5)
Results of weighted least-square fit	$\gamma=1.25\pm0.015$ including systematic errors				$\gamma=1.25\pm0.04$ including systematic errors				$\gamma=1.18\pm0.04$ including systematic errors				$\gamma=1.23\pm0.03$ including systematic errors				Results of least-squares fit				ν: 0.64 ± 0.02; k: 2.93 ± 0.31	0.65 ± 0.02; 2.99 ± 0.32

[a] Beam-monitor preset count.

average count rate per beam-monitor preset count (BMpc as indicated for the background) at a superlattice reflection resulting from n repetitions. I_0 is the unfolded critical peak intensity, i.e.,

$$I_0=(\langle I\rangle-B)/\mathrm{IR}(\kappa_1(\Delta T/T)),$$

where IR is the peak intensity reduction due to resolution.[1] A weighted least-squares fit of $I_0\propto(\Delta T/T)^{-\gamma}$ gave the value of γ, and the number in parenthesis following I_0 is the deviation in % between I_0 and the least-squares fit. The weight w on each point is proportional to the inverse square of the resulting uncertainty on I_0, i.e.,

$$w=\left\{\frac{\langle I\rangle I_0^2}{n(\langle I\rangle-B)^2}+I_0^2\left(1.25\frac{\delta T}{\Delta T}\right)^2\right\}^{-1}.$$

Here δT is the uncertainty on ΔT; we estimated $\delta T\approx 0.1°\mathrm{C}$. It should be noted that if γ is determined from a linear least-squares fit of $\log I_0$ versus $\log(\Delta T/T)$ the weight in this fit is wI_0^2.

Inverse Correlation Range $a\kappa_1$

κ_1 is the inverse correlation range parameter in the Ornstein-Zernike correlation function $e^{-\kappa_1 r}/r$. a is the cube edge of the unit cell (2.97 Å). $a\kappa_1$ is thus dimensionless. In Type of scan, x means scan along $\boldsymbol{\tau}_0$ and y means scan perpendicular to $\boldsymbol{\tau}_0$ in the scattering plane. A least-squares fit of $a\kappa_1=k(\Delta T/T_c)^{\nu}$ gave the values of ν and k, and the number in parenthesis following $(a\kappa_1)$ is the deviation in % between $a\kappa_1$ and the least-squares fit. The values of k and ν were used to find $\mathrm{IR}(\kappa_1(\Delta T/T))$ in the evaluation of I_0.

PHYSICAL REVIEW VOLUME 153, NUMBER 3 15 JANUARY 1967

Long-Range Order and Critical Scattering of Neutrons below the Transition Temperature in β-Brass

J. ALS-NIELSEN AND O. W. DIETRICH
The Danish Atomic Energy Commission, Research Establishment Risö, Roskilde, Denmark
(Received 11 July 1966)

The temperature dependence of long-range order $\langle P_o\rangle$ has been determined from the temperature variation of a superlattice Bragg reflection. The results fitted a power law $\langle P_o\rangle\propto(T_c-T)^{\beta}$ with T_c the critical temperature and $\beta=0.305\pm0.005$, in agreement with the theoretical prediction $0.303<\beta<0.318$ obtained from a comparison of the measured line profiles of the (1,0,0) superlattice reflection at 15.5 and 2.2° below T_c. The ratio between the susceptibilities $\chi^+(\Delta T)$ and $\chi^-(\Delta T)$ ΔT deg below and above T_c was found to be 5.5 ± 2.0 at $\Delta T=4.4°$ and 8.5 ± 3.0 at $\Delta T=2.1°$. The Ising-model theory predicts the ratio to be 5.2 independent of ΔT.

INTRODUCTION

IN the ordered state in β-brass the Cu and Zn atoms share the sites in the bcc lattice, so Cu atoms predominantly occupy, say, the cube corners and Zn atoms the cube centers of the unit cells. The occupation of lattice site $\mathbf{l}$ is given by the occupation variable $S_{\mathbf{l}}$, which is $+1$ if site $\mathbf{l}$ is occupied by a Cu atom and -1 if site $\mathbf{l}$ is occupied by a Zn atom.

The long-range order $\langle P_o\rangle$ is defined as the average of the occupation variable throughout the lattice on the corner sites or the center sites, or the *weighted* average occupation of *all* lattice sites ascribing a weight factor of $+1$ to the corner sites and -1 to the center sites. The weight factor is conveniently expressed by $e^{i\boldsymbol{\tau}\cdot\mathbf{l}}$, where $\boldsymbol{\tau}$ is any vector in reciprocal space with an odd sum of indices (a reciprocal superlattice vector). Thus

$$\langle P_o\rangle\equiv\frac{1}{N}\sum_{\mathbf{l}}S_{\mathbf{l}}e^{i\boldsymbol{\tau}\cdot\mathbf{l}}\equiv\frac{1}{N}\sum_{\mathbf{l}}P_{\mathbf{l}}. \quad (1)$$

The short-range order is described by the correlation of the occupation of two lattice sites separated by the lattice vector $\mathbf{r}$. However, to distinguish short-range order from long-range order, it is only the asymptotically vanishing part $p(\mathbf{r})$ of the pair-correlation function that describes the short-range order, i.e.,

$$p(\mathbf{r})\equiv\langle P_oP_{\mathbf{r}}\rangle-\langle P_o\rangle^2, \quad (2)$$

where

$$\langle P_oP_{\mathbf{r}}\rangle\equiv\frac{1}{N}\sum_{\mathbf{l}}P_{\mathbf{l}}P_{\mathbf{l}+\mathbf{r}}. \quad (3)$$

Note that $p(\mathbf{r})$ is zero both in the completely ordered state and in the completely disordered state.

When the temperature increases, the long-range order gradually decreases and finally vanishes at the critical temperature T_c. However, the correlation range tends to infinity when T approaches T_c. It is possible to study these phenomena experimentally by means of neutron diffraction. The differential scattering cross section $d\sigma/d\Omega$ at scattering vector $\boldsymbol{\kappa}$ is given by[1]

$$\frac{d\sigma}{d\Omega}\propto N\langle P_o\rangle^2\delta(\boldsymbol{\kappa}-\boldsymbol{\tau})+\sum_{\mathbf{r}}p(\mathbf{r})e^{i(\boldsymbol{\kappa}-\boldsymbol{\tau})\cdot\mathbf{r}}. \quad (4)$$

[1] J. Als-Nielsen and O. W. Dietrich, second preceding paper, Phys. Rev. 153, 706 (1966).