

STRUCTURE AND THERMODYNAMIC PROPERTIES OF CRYOCRYSTALS

Handbook

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Handbook

V. G. Manzhelii

*Professor, Doctor in Physics and Mathematics
Head of Department of Thermal Properties of Molecular Crystals
Low Temperature Physics and Engineering Institute
Ukrainian Academy of Sciences*

A. I. Prokhvatilov

*Leading Research Fellow
Doctor in Physics and mathematics*

V. G. Gavrilko

*Senior Research Fellow
Ph. Doctor in Physics and mathematics*

A. P. Isakina

*Senior Research Fellow
Ph. Doctor in Physics and mathematics*

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Preface

This handbook includes data on the structures of phases, diagrams of phase equilibria, and physical properties of simplest molecular crystals (cryocrystals) such as nitrogen, carbon monoxide, oxygen, fluorine, carbon dioxide, nitrogen monoxide, hydrogen sulfide, methane, and deuterated methanes. The data are presented in the form of tables and graphs; the most reliable results are noted and their accuracy is indicated where possible.

The handbook is intended for a wide circle of specialists that use simple molecular crystals in their investigations and practical activity. This book is also of interest for scientists, postgraduates, and students that are specialists in the field of molecular physics and low-temperature physics and technology.

Foreword

The simple van der Waals solids that crystallize at low temperatures are called cryocrystals in the recent years. They include monatomic cryocrystals of neon, argon, krypton, and xenon, and simple molecular crystals with various types of noncentral intermolecular interaction, such as hydrogen, nitrogen, oxygen, fluorine, carbon monoxide and dioxide, nitrogen monoxide (quadrupole interaction); methane, silane, and their deuterated forms (octupole interaction); ammonia, hydrogen sulfide (dipole interaction, hydrogen bonds), etc. Because of the relative simplicity of these substances, they can easily be considered theoretically and are frequently used as model objects for solving fundamental problems of solid state physics such as lattice dynamics, phase transformations, impurity effects, etc.

Cryocrystals also include a group of substances with special properties – quantum crystals of helium and hydrogen – in which the zero-point energy of their molecules is comparable with the energy of intermolecular interaction. Quantum properties are also exhibited by the rotational subsystem of solid methane. Unique properties are also characteristic of solid oxygen, which combines the properties of a molecular crystal and a magnetic material.

In recent years, cryocrystals find growing practical application in the cryogenic, space, laser, high-pressure technologies, nuclear physics, and other fields of science and technology.

By the time of this writing, a vast body of experimental data on the physical properties of cryocrystals were accumulated, which were published not only in original papers, but also in numerous textbooks and handbooks. The most com-

prehensive report on the properties of monoatomic cryocrystals was given in Klein, M. L., and Venables, J. A., (Eds.), *Rare Gas Solids*, London, Academic, vol. 1 (1976) and vol. 2 (1977); the properties of molecular cryocrystals were considered in most detail in the textbooks of Verkin, B. I., and Prikhot'ko, A. F., (Eds.), *Cryocrystals*, Kiev, Naukova Dumka (1983) and Manzhelii, V. G., and Freiman, Yu. A., (Eds.), *Physics of Cryocrystals*, New York, AIP (1996). Special handbooks were devoted to individual substances, e.g., Roder, H. M., et al., *Survey of the Properties of the Hydrogen Isotopes below Their Critical Temperatures*, NBS, Technical Note 641, U. S. Government Printing Office, Washington, DC (1973); Eselson, B. N., et al., *Svoistva zhidkogo i tverdogo geliya (Properties of Liquid and Solid Helium)*, Kiev, Naukova Dumka (1978); Sauer, P. S., *Cryogenics Hydrogen Data Pertinent to Magnetic Fusion Energy*, Lawrence Livermore Laboratory (1979); Eselson, B. N., et al., *Svoistva zhidkogo i tverdogo geliya. Rastvory ^3He – ^4He (Properties of Liquid and Solid Helium: ^3He – ^4He Solutions)*, Kiev, Naukova Dumka (1982); Verkin, B. I., et al., *Properties of Condensed Phases of Hydrogen and Oxygen*, New York, Hemisphere (1990).

With the above in mind, we devoted this handbook to those cryocrystals that, although they are of a significant research and applied interest, have not yet been described in sufficient detail in modern reference books.

In this handbook, we analyze and systematize the results of studies on the structures, phase diagrams, P – V – T data and thermophysical properties (thermal expansion, heat capacity, thermal conductivity, heats of phase transitions, vapor pressures, compressibilities) of cryocrystals such as nitrogen, carbon monoxide and dioxide, oxygen, fluorine, nitrogen monoxide, hydrogen sulfide, methane, and deuterated methanes. Some principal information on the molecules and properties of substances considered in this handbook are given in Table 1.

When compiling this handbook, we partly took into account materials that were published in the first two monographs mentioned above, but in the major cases we used the data of original works. Quantitative experimental data are mainly represented in the form of tables, and the general dependences of physical properties on thermodynamic parameters are shown in figures. The values that are given in tables represent either generalized experimental data of several investigations or the results of a single most detailed and reliable work. In those cases where the accuracy of measurements is not sufficient or no tabulated data are given in the original work, we only give experimental data in the form of graphs. In those cases where no preference could be given to any of the works available, the tables contain values obtained by different authors. When analyzing the results for some substances (nitrogen, hydrogen sulfide, methane), large attention was paid to isotopic effects, which are of special interest from the physical viewpoint.

Table 1 Principal physical parameters of simple molecules and cryocrystals

Parameter	$^{14}\text{N}_2$	$^{15}\text{N}_2$	CO	O ₂	F ₂	N ₂ O	CO ₂	CH ₄	CD ₄
Molecular weight M , a.m.u.	28.0134	30.03073	28.01055	31.9988	37.9968	44.0128	44.00995	16.04303	20.07491
Molecule symmetry	$D_{\infty h}$	$D_{\infty h}$	$C_{\infty v}$	$D_{\infty h}$	$D_{\infty h}$	$C_{\infty v}$	$D_{\infty h}$	T_d	T_d
Equilibrium bond length, Å	1.0977	1.0977	1.1283	1.207	1.4177	N-N 1.1282 N-O 1.1842	1.1599	C-H 1.1014 H-H 1.803	C-D 1.0986 D-D 1.801
Dipole moment D , 10^{-18} esu	-	-	0.112	-	-	0.166	-	-	-
Quadrupole moment Q , 10^{-26} esu	-1.4	-	-2.5	-0.39	-0.88	-3.0	-4.3	-	-
Octupole moment I , 10^{-34} esu	-	-	-	-	-	-	-	+4.5	+4.3
Averaged polarizability α , Å	1.767	-	1.977	1.60	-	3.0	2.63	2.60	2.60
Anisotropy of polarizability k	0.1313	-	0.0897	0.2375	-	0.329	0.2664	-	-
Rotational constant B , K	2.8751	2.6840	2.7787	2.081	1.27	0.60592	0.56355	7.56	1.89
Parameters of the 6-12 Lennard-Johnes potential									
ϵ , K	91.5	91.5	110.0	125.0	116.0	220.0	190.0	148.2	148.2
σ , Å	3.681	3.681	3.590	3.136	3.153	3.879	3.996	3.817	3.817
Nearest-neighbor spacing at $T = 0$ K, r_0 , K	3.994	3.990	3.993	3.134	2.979	3.988	3.927	5.858	5.768
Debye temperature at $T = 0$ K, Θ_D , K	83.6	81.3	103.3	104.5	110.0	141.0	151.8	141.0	138.0

Nomenclature

a, b, c, β'	lattice parameters
B_T	isothermal modulus of elasticity
C_{ij}	elastic constants
C_P	heat capacity at constant pressure
C_S	heat capacity at saturated vapor pressure
ΔH	heat of phase transition
L_0	heat of sublimation at $T = 0$ K
L_m	latent heat of fusion
M	molecular weight
$P; P_m$	pressure; pressure at the triple point
R	universal gas constant
ΔS_m	entropy change in melting
ΔS_{subl}	entropy change in sublimation
T_m	triple-point temperature
V	molar volume
V_s, V_l	molar volume of solid and liquid phases
V_m^s, V_m^l	molar volume of solid and liquid phases at the melting curve
α, β	linear and bulk thermal expansion coefficients
Θ_D	Debye temperature
α_S, α_T	adiabatic and isothermal compressibilities
λ	thermal conductivity
ρ	density
χ	magnetic susceptibility
v_l, v_t	longitudinal and transverse sound velocities

1

Nitrogen*

1.1 Phase Equilibria

1.1.1 Phase diagram

In spite of the simplicity of the N_2 molecule and interaction of intermolecular forces, the equilibrium phase diagram P - T of nitrogen is complicated and it took efforts of many researchers in the past 80 years to establish it. The majority of these works were comprehensively analyzed in the reviews by Eters (1988), Freiman (1990) and in the book by Manzhelii and Freiman (1996). Table 1.1 lists experimental works up to 1995, in which the phase diagram of nitrogen is studied by various methods. The last column of Table 1.1 presents the major results of these works related to the phase diagram. The P - T phase diagram is shown in Figure 1.1 and P - V - T data are presented in Figure 1.2. Apart from α and β phases existing at the equilibrium vapor pressure, five more phases are found at high pressures γ , δ , ϵ , ζ , and η . Moreover, Reichlin et al. (1985) obtained evidence of additional phase transitions into phases θ and ι (Figure 1.1) at room

*Hereafter, the term "nitrogen" and symbol " N_2 " are used to denote nitrogen-14 isotope with the natural content of nitrogen-15. The data for nitrogen-15 are otherwise specified.

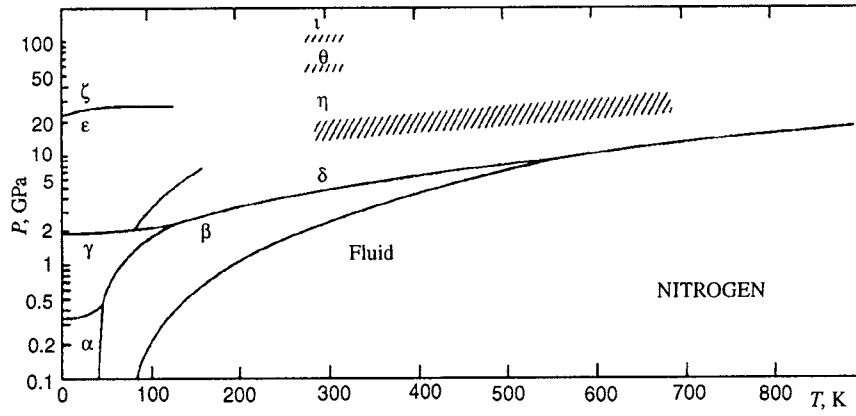


Fig. 1.1 The P - T phase diagram of nitrogen (Schiferl et al., 1988).

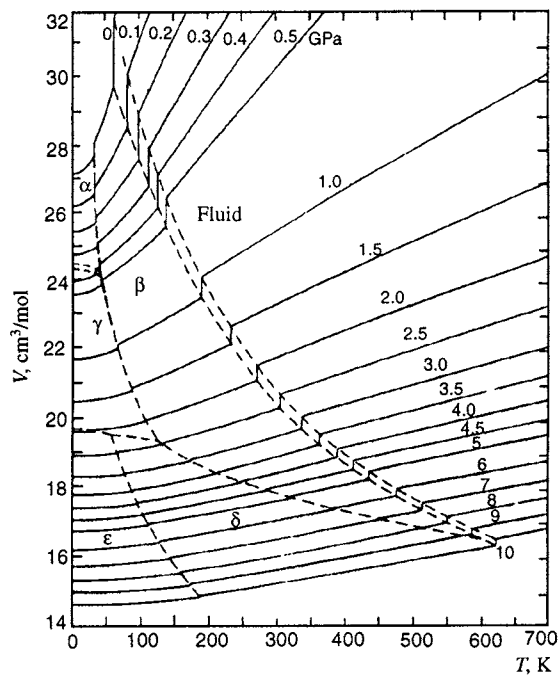


Fig. 1.2 The V - T diagram of nitrogen (Mills et al., 1986). The dotted lines correspond to the phase boundaries, the solid lines to isobars which demonstrate jumps in the volume upon phase transitions.

Table 1.1 Sources of data on the phase diagram of nitrogen

No	Type of measurement	Pressure range (kbar)	Temperature range (K)	References	Comments
1	Heat capacity	0	20-60	Eucken (1916); Clusius (1929); Giauque and Clayton (1933)	Discovering and studing of α - β phase transition
2	Piston displ.	0-10	4.2-53	Swenson (1955)	Discovering of γ - N_2 ; measurements of α - γ , α - β , β - γ transition lines; 4.2, 39.5 and 53 K isotherms
3	Piston displ.	0-19	65	Stewart (1956)	65 K isotherm
4	X-ray	4	20.5	Mills and Schuch (1969)	Crystal structure of γ - N_2 .
5	X-ray	0-4	20-49	Schuch and Mills (1970)	Lattice constants of α , β , γ phases
6	NQR	0-3.3	35.6-44.5	Brookeman and Scott (1973)	α - β transition line
7	Piston displ.	16-20	247; 273	Mills et al. (1975)	247 and 273 K isotherms
8	X-ray	25.1	296	Schiferl et al. (1978)	Lattice parameters of β -phase
9	Raman	up to 374	300	LeSar et al. (1979)	Discovering of δ -phase
10	X-ray	49	293	Cromer et al. (1981)	Crystal structure of δ -phase
11	X-ray	29.4	300	Schiferl et al. (1983)	Lattice parameters of β -phase
12	Raman	4-52	15-300	Buchsbaum et al. (1984)	β - δ and γ - δ transition lines
13	X-ray	50-10	296	Olinger (1984)	Compressibility and lattice parameters along 296 K isotherm

Table 1.1 Sources of data on the phase diagram of nitrogen (Continued)

No	Type of measurement	Pressure range (kbar)	Temperature range (K)	References	Comments
14	Raman	4–270	15	Schiferl et al. (1985)	Discovering of low temperature ϵ and ζ phases
15	Raman	up to 1300	300	Reichlin et al. (1985)	Three new high pressure phase transitions at 200, 660 and 1000 kbars, and weakening of the intramolecular N–N bond
16	Brillouing scattering	0–210	300	Grimsditch (1985)	δ - η phase transition at 200 kbar
17	X-ray	40–130	100–300	Mills et al. (1986)	Phase diagram; crystal structure of ϵ and ζ phases; pressure and temperature dependences of lattice parameters
18	Raman	up to 1800	300	Bell et al. (1986)	Weakening of the intramolecular N–N bond
19	Raman	23–180	290–900	Zinn et al. (1987)	Phase diagram; β - δ phase boundary
20	X-ray	up to 650	300	Jephcoat et al. (1988)	A sequence of structure phase transition
21	Isochoric scanning	up to 85	150–550	Vos and Schouten (1990)	Solid-solid-fluid triple point; δ - β transition and melting line
22	X-ray	57.5–439	300	Olijnyk (1990)	δ - ϵ transition line, P-V data and lattice parameters
23	Raman	up to 180	up to 900	Schmidt et al. (1991)	Revised data Zinn et al. (1987)
24	Raman	80–540	300	Schneider et al. (1992)	A new phase transition above 40 GPa
25	Raman	42–127	120–400	Schreeboom and Schouten (1993)	A new phase transition between δ and ϵ phases

Table 1.2 Equilibrium triple points on the P - T diagram of nitrogen

Triple point	T , K	P , GPa	Source
β - δ -fluid	580	9.9 ± 0.5	Young et al. (1987)
	578 ± 10	9.9 ± 0.5	Zinn et al. (1987)
	555 ± 5	8.0 ± 0.2	Vos and Schouten (1989)
α - β - γ	44.5	0.465	Schuch and Mills (1970)
	46.0	0.460	Brookeman and Scott (1973)
γ - δ - ϵ	70.0	2.10	Young et al. (1987)
γ - β - δ	130.0	2.40	Young et al. (1987)

temperature and pressures around 66 and 100 GPa. Spectroscopy investigations by Bell et al. (1986) and Reichlin et al. (1985) at 100, 130, and 180 GPa revealed the weakening of intramolecular N-N bonds, which suggests "metallization" of nitrogen. According to the experimental data available now, the diagram of nitrogen contains three equilibrium triple points of solid phases α - β - γ , γ - δ - ϵ , and γ - β - δ , one equilibrium triple point of two solid phases with liquid β - δ -fluid, and a triple point at $P = 0$ for the equilibrium of the solid phase with liquid and vapor β -fluid-vapor (Figures 1.1 and 1.2). The coordinates of the triple points are listed in Table 1.2.

1.1.2 Phase Transitions and Structure of Phases

Low-Pressure Phases at $P = 0$

The phase transition α - β at the equilibrium vapor pressure was first found by Eucken (1916) and its existence was later confirmed by Clusius (1929), Giaque and Clayton (1933) in their studies on heat capacity of solid nitrogen. The basic characteristics of the α - β transition are presented in Table 1.3. The values of an jump in the molar volume derived from dilatometric measurements by Manzhelii et al. (1966) and Heberlein et al. (1970) differ radically from the values obtained by X-ray diffraction (Krupskii et al., 1975) and the compressibility methods (Swenson, 1955). The data of the latter works seem more reliable. The crystal structure of the α and β phases was studied by many authors, and these works are enumerated in Table 1.4.

The α -phase was first reliably studied by X-ray diffraction method by Ruheman (1932). Its orientational structure was assumed to have the centrosymmetric space group of the $Pa3 (T_h^6)$ symmetry. The unit cell contains 4 mole-

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