Structural characteristics of solid nitrogen. Isotopic effect

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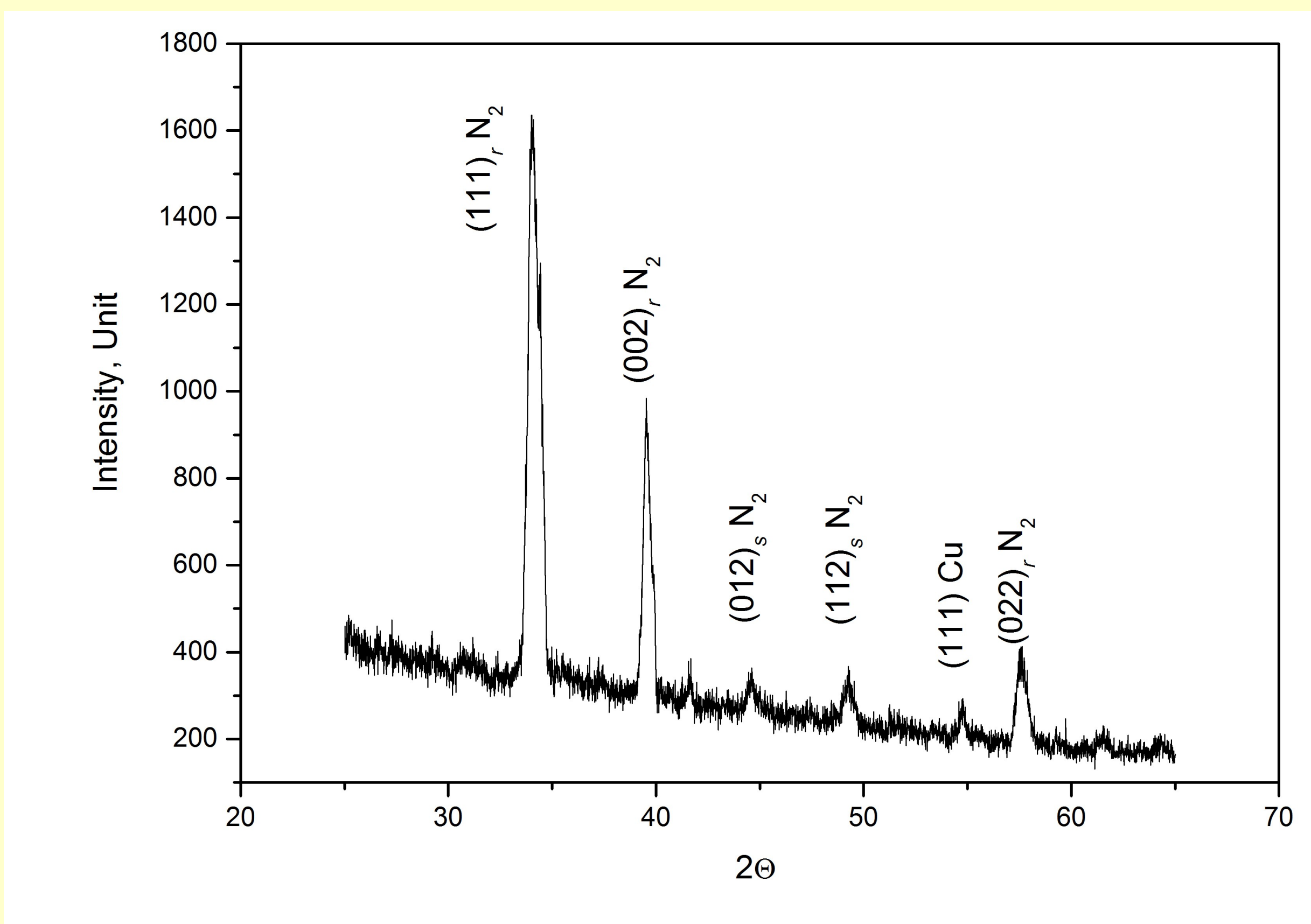*e-mail: aksenova@karazin.ua*

Fig. 1. Typical powder x-ray patterns for ordered N₂ phase at 5 K, "regular" (r) and "superstructure" (s) denotes reflections for *Pa3* lattice
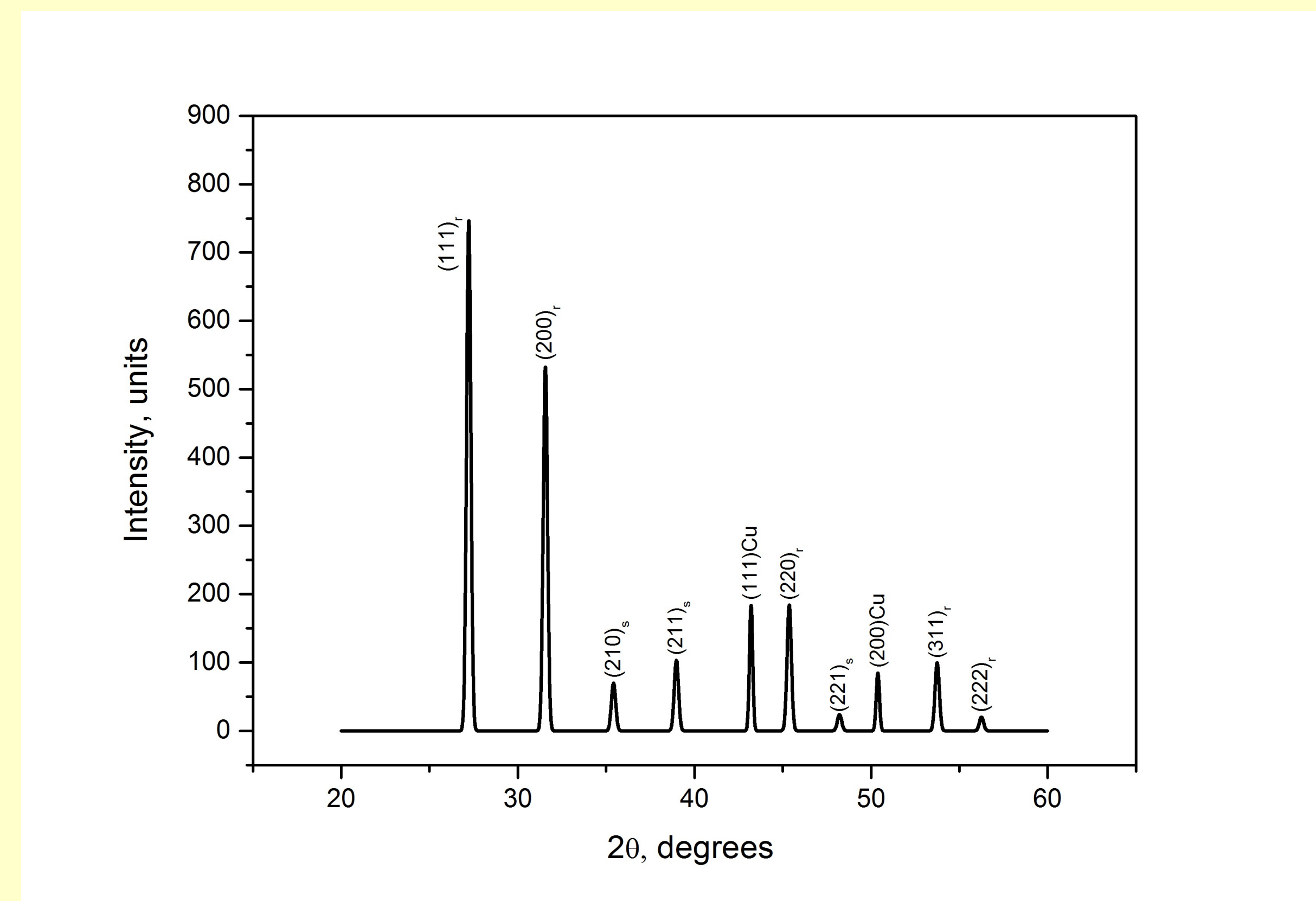

Fig. 2. Typical powder x-ray pattern for $^{15}N_2$ ordered phase at 7 K, "regular" (*r*) and "superstructure" (*s*) denotes reflections for *Pa3* lattice (lattice parameter *a*=5.649 Å with 4 molecules per unit cell)
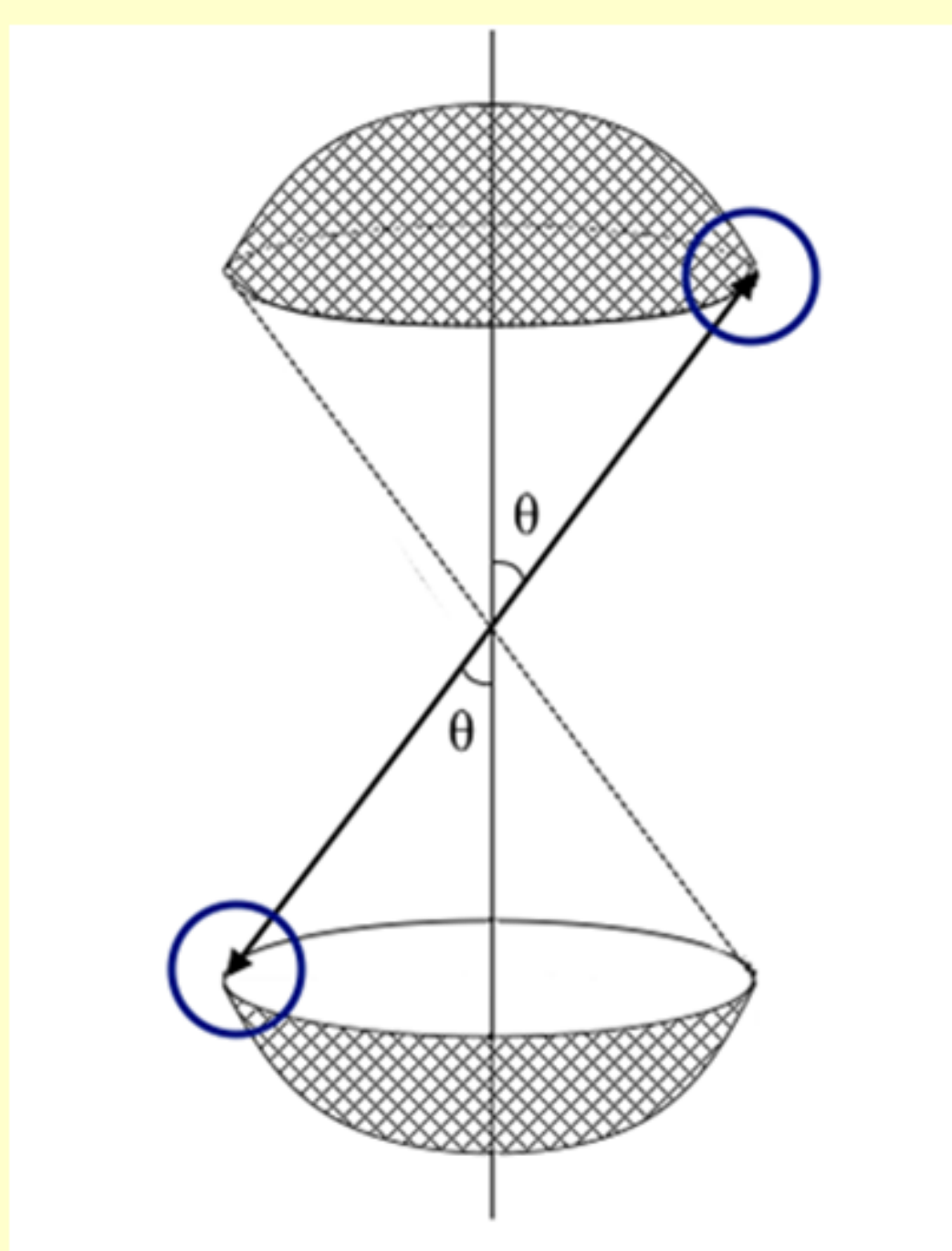

Fig. 3. Schematic image of the librations for diatomic molecule. The shaded area is the surface on which the atom is uniformly «smeared»

The isotopic effect qualitatively manifests on X-ray diffractograms in the displacement of characteristic diffraction peaks due to the difference in molar volumes of isotopes (Fig. 1 and 2).

Solid nitrogen has been selected as the research object. Nitrogen has two stable isotopes: nitrogen-15 ($^{15}N_2$) and nitrogen-14 ($^{14}N_2$).

The diffraction pattern of nitrogen in the low-temperature phase can be represented as "regular" and "superstructural" reflections (Fig. 1 and 2).

Both isotopes throughout the solid phase domain have a phase transition from a primitive cubic lattice to a hexagonal lattice. The peculiarity of this transition is that along with the slippage of crystallographic planes in the crystal, with the temperature the rotational motion from librations to the free rotation of the linear nitrogen molecule changes (Fig.3).

The aim of our work is to see how the isotope composition affects the X-ray reflection in a molecular crystal.

$$I \propto |F|^2$$ Intensity x-ray reflection

$$F(\vec{S}) = F_r(\vec{S}) + F_s(\vec{S})$$ Scattering crystal cell Fig. 4

$$F_r(\vec{S}) = 8f(\vec{S})j_0(\xi S)e^{-M(S)}$$ Scattering sublattice on Fig. 4b for regular reflection

$$F(\vec{S}) = 10f(\vec{S})\eta j_2(\xi S)G_c(\vec{S})e^{-M(S)}$$ Scattering sublattice on Fig. 4c for super-structural reflection

$$e^{-M(S)}$$ Debye-Waller temperature factor

$$f(\vec{S})$$ Atomic scattering factor of nitrogen

$$f(\vec{S})j_0(\xi S)$$ and $$f(\vec{S})\eta j_2(\xi S)$$ electron density distribution at sublattice sites Fig. 4 b and c
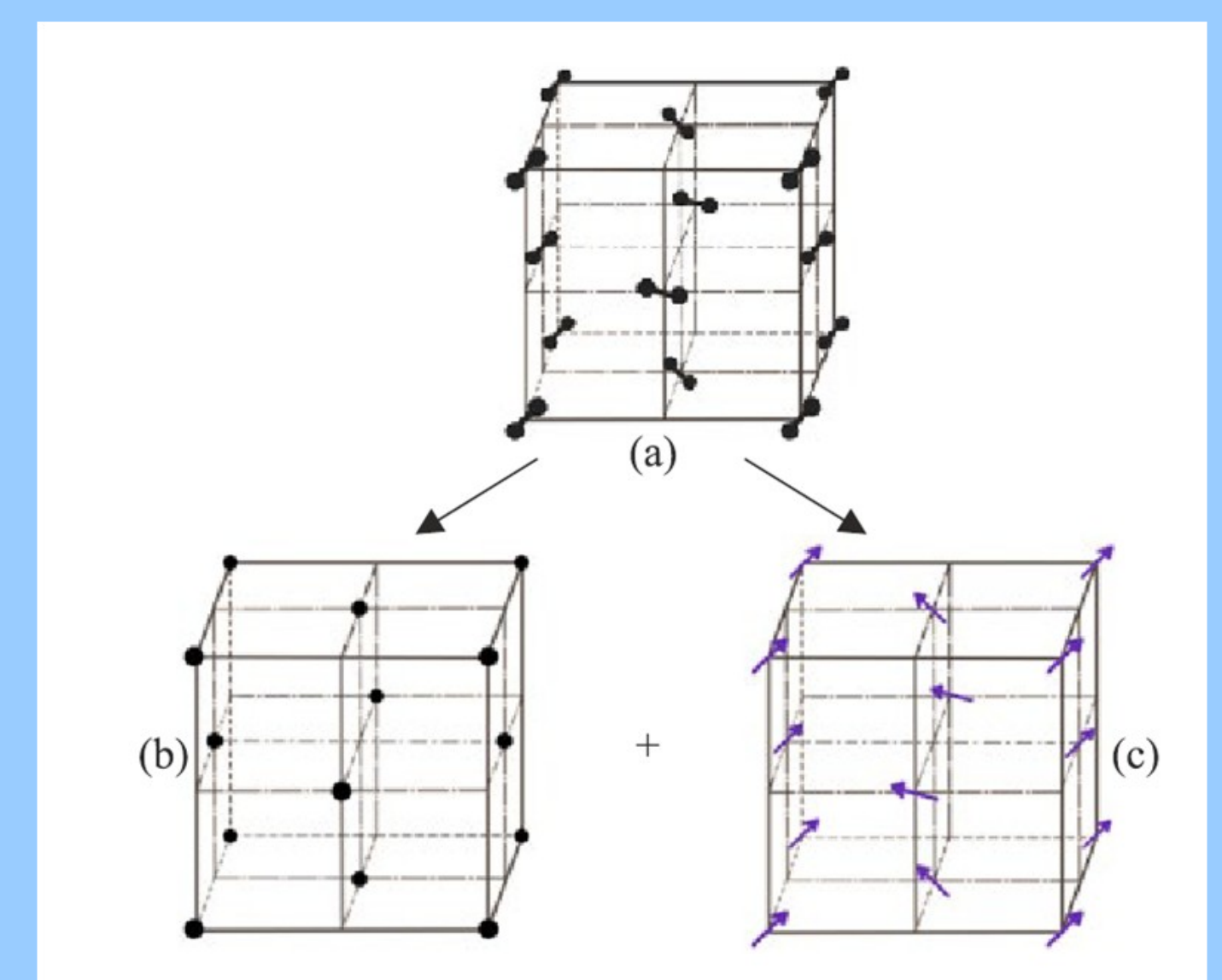

Fig. 4. The lattice of solid N₂ in ordered phase is *Pa3* structure (a), and representing itself, as a two sublattices, where the structure is *fcc* (b) and 4 primitive cells after shifted by half diagonal relatively to each other (111) (c).
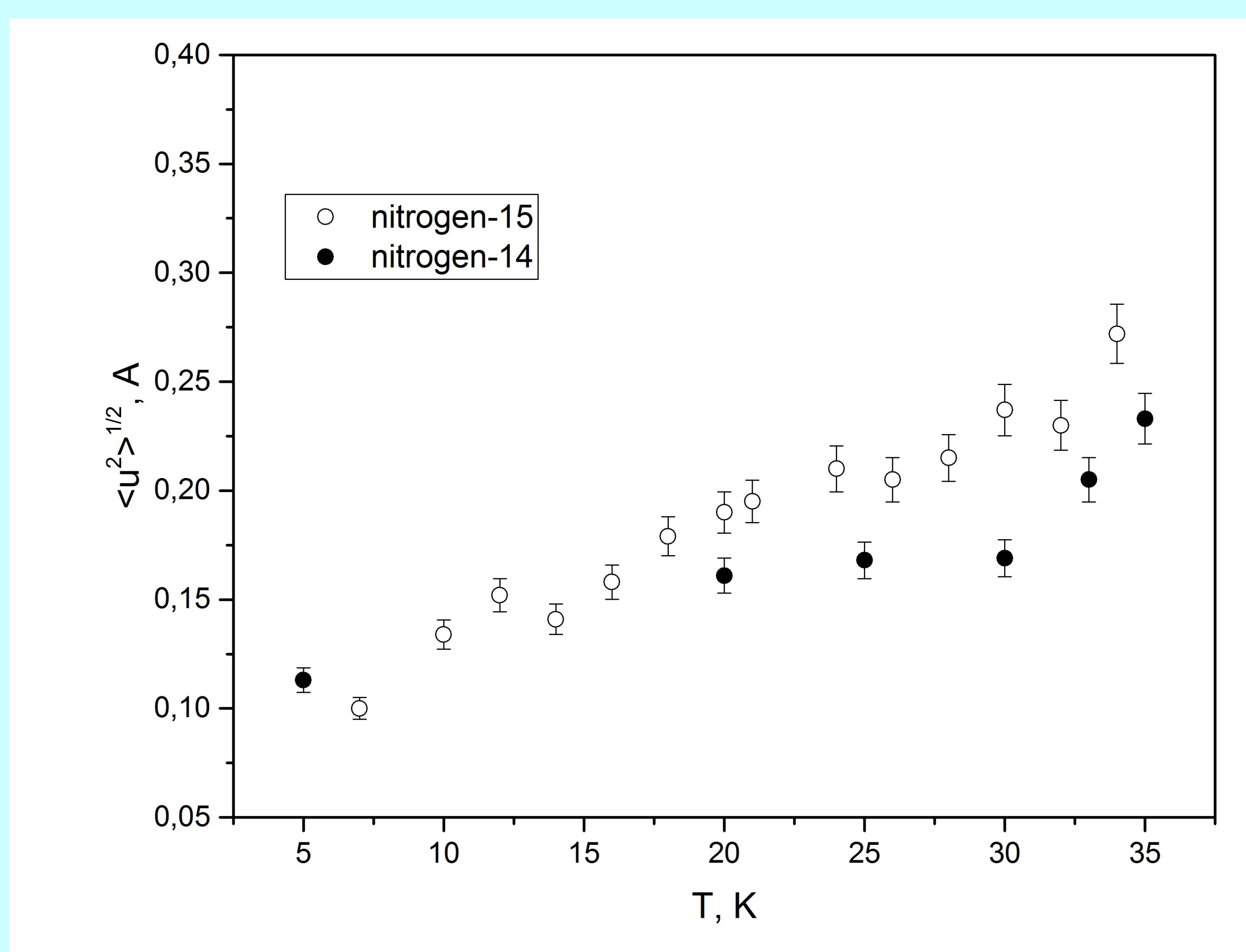

Fig. 5. The mean square displacement of molecules from lattice point in nitrogens as a function of temperature.
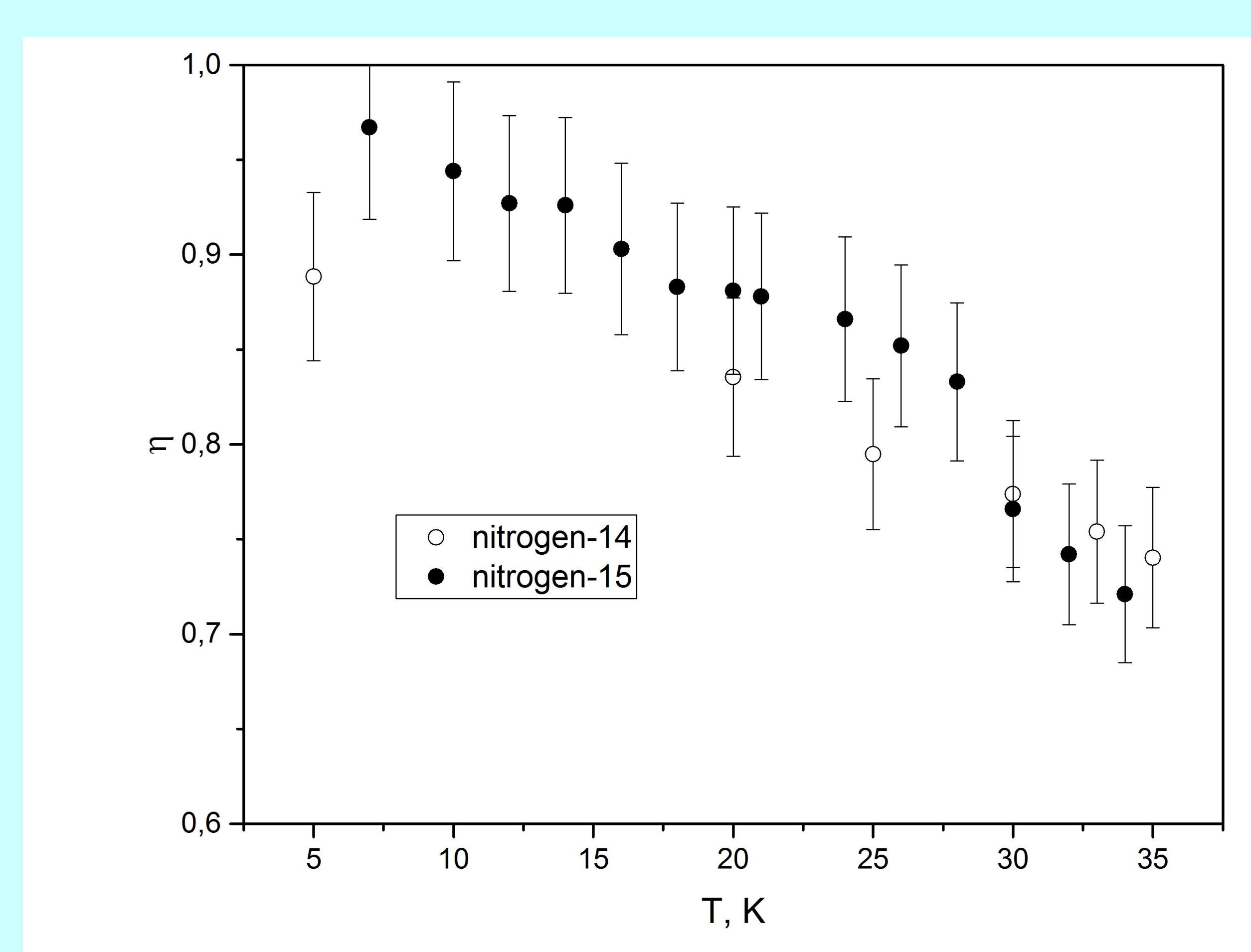

Fig. 6. Orientational order parameter as a function of temperature for solid nitrogens

The analysis of the intensity for regular X-rays (Fig. 1, 2) was constructed temperature dependence of the displacement for the centers mass of linear molecules from the lattice site (Fig. 5). It can be seen that the difference in molecular masses begins to appear above 20K. From the analysis of the all intensities given in Fig. 1 and 2 were built temperature dependencies of the orientation parameter for solid nitrogen 14 and 15 (present on Fig. 6).

It should be noted that for both linear molecule nitrogen isotopes at 0K perform libration ostillatoins. The effect of mass difference on the nitrogen molecule in the low-temperature phase is observed at hydrogen temperatures (T below 10K).