Highlights in Functional Materials Research

Vienna, 15 – 16 Feb 2018

Colloquium celebrating the birthdays of Gerhard Krexner and **Romano Rupp**

Topics

The colloquium covers topics related to the research work of our colleagues Gerhard Krexner and Romano Rupp. These are:

Neutron scattering, Neutron holography, hydrogen in metals, martensitic phase transformations, etc.

Photonic crystals, nonlinear optic response of photosensitive functional materials, optical holography, etc.

Location

Faculty of Physics of the University of Vienna

Boltzmanngasse 5, A-1090 Wien

Ernst-Mach Hörsaal, 2nd floor

Organized by the group

Physics of Functional Materials

Secretary: Sabine E. Vranckx Herrera

Questions: wilfried.schranz@univie.ac.at

Invited speakers

Pierre Tolédano
University de Picardie, Amiens
France

Jan Lagerwall
University of Luxembourg,
Luxembourg

Michael Zehetbauer
University of Vienna, Austria

Serguey Odoulov
National Academy of
Sciences of Ukraine,
Ukraine

Götz Eckold
Georg-August Universität
Göttingen, Germany

Xinzheng Zhang
Nankai University , Tianjin,
China

Michael Jentschel
Institut Laue-Langevin, Grenoble,
France

Program

Time/Chair	**Thursday** 15.2.2018	Time/Chair	**Friday** 16.2.2018
10:00 – 10:30	Opening	10:00 – 10:30 W.Schranz	G. Krexner
10:30 – 11:00 M. Fally	R. Rupp	10:30 – 11:15 G. Krexner	P. Tolédano – Physical properties of multiferroic crystals under illumination
11:00 – 11:30	Coffee break	11:15 – 11:45	Coffee break
11:30 – 12:15 R. Rupp	J. Lagerwall - Cholesteric liquid crystal shells: from unique photonic crystal properties to applications in non-biometric secure authentication tags	11:45 – 12:30	M. Zehetbauer - Optimizing Functional Nanostructured SolidsThrough Lattice Defect Engineering by Severe Plastic Deformation
12:15 – 14:00	Lunch	12:30 – 14:15	Lunch
14:00 – 14:45 I. Drevenšek-Olenik	S. Odoulov – $Sn_2P_2S_6$ - A curious nonlinear optical material	14:15 – 15:00 J. Klepp	M. Jentschel - Gamma Ray Spectroscopy at the Institut Laue-Langevin
14:45 – 15:30	X. Zhang - Surface Plasmons in Various Graphene Structures	15:00 – 15:45	G. Eckold – $SrTiO_3$, well known, but still surprising
15:30 – 16:00	Coffee break	15:45 – 16:00	Closing
16:00 – 18:00	Poster Session		
19:00	Conference Dinner – Wiener Rathauskeller*		

*Invited by the mayor of the city of Vienna Dr. Michael Häupl

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Cholesteric liquid crystal shells: from unique photonic crystal properties to applications in non-biometric secure authentication tags

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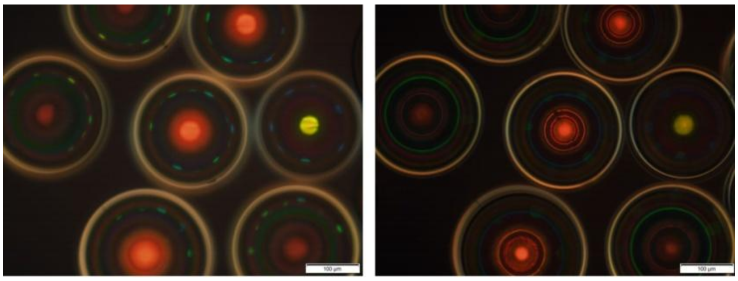
The colorful circularly polarized iridescence of short-pitch cholesteric liquid crystals (CLCs), arising due to Bragg reflection by the periodic internal helix structure, has fascinated scientists and laymen alike for more than a century. Recently, the peculiar effects arising when cholesterics are confined in samples with curved interfaces, such as droplets, shells and fibers, have received increasing attention [1]. Cholesteric shells are particularly interesting thanks to the rich optics arising when the axis of periodicity rotates continuously around the shell, with the added option of varying the shell thickness between top and bottom. The shell reflects light straight back to the observer regardless of viewing direction, and if a cluster of cholesteric shells in a plane is illuminated by white light, an intricate photonic cross communication pattern arises (left photo), with colourful spots arranged in a symmetry that reflects the arrangement of the shells [2-3]. If the shell is asymmetric with a thin top, light enters into the shell interior. The CLC inside then acts as a selective optical echo chamber that creates a colourful pattern of concentric rings (right photo) [4]. By changing the focus one can accentuate the internal reflection ring pattern or the external cross communication pattern, which is still present. The shells can be made durable and mechanically robust by polymerizing or polymer-stabilizing them, making large-scale application of the shells viable.
For instance, by incorporating a cluster of CLC shells within an index matched solid matrix, a tag can be produced that generates the CLC shell patterns upon illumination, changing dynamically in response to which sample area is illuminated, to the focus, to the angle of illumination, and to the polarization and spectral content of the illuminating light. Moreover, since the spheres are arranged in a random fashion each tag is unique in a way that is out of control of the producer: not even the manufacturer of the original would be able to make a copy. In an interdisciplinary research thrust at the University of Luxembourg, involving computer and materials scientists, we are exploring the potential of using such CLC shell-based tags for secure authentication [5]. The tags could identify persons or be integrated in objects prone to counterfeiting, to prove the authenticity of the original product.



Left: Patterns arising from communication between cholesteric shells, mediated by reflections on the sphere outsides. Right: A change in focus reveals that light enters through the thin top of these asymmetric shells, giving rise to different pattern of concentric colourful rings.

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$Sn_2P_2S_6$ – A curious nonlinear optical material

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Nearly everything with tin hypothiodiphosphate ($Sn_2P_2S_6$) synthesized in Darmstadt in 1974 is unusual. It is one of rare ferroelectric materials that allows for observing and studying tricritical Lifshitz point not far away from ambient temperature. Being optically biaxial it brings rare opportunity to study in details the conical refraction. Various nonlinear effects are quite pronounced, too. The largest of 10 nonvanishing Pockels coefficients, for example, is more than 20 times bigger than that of a standard EO crystal $LiNbO_3$. Among other arguments, this one motivated Dr. Romano RUPP with his colleagues to start a systematic study of $Sn_2P_2S_6$ photorefractive properties in 1995 [1]. Even the first measurements brought a nice result that could hardly be expected for large bandgap crystal with no deliberately introduced extrinsic defects – material manifested photorefractivity in the near infrared (1.06 μm). The fast temporal response, in the millisecond range, appeared to be the other property quite different from that expected for ferroelectric photorefractives (like $BaTiO_3$, $LiNbO_3$ and others).

The decades of R&D efforts in Germany, Ukraine, Switzerland, France, Austria, China, Japan, USA and UK revealed a lot of new particular properties of material itself and also many new manifestations of these particularities in nonlinear optical interactions and applications. The nonlinear coupling constant of $Sn_2P_2S_6$ was considerably increased with deliberate doping while its spectral sensitivity was extended to optical communication wavelength 1.5 μm. The efficient dynamic phase conjugate mirrors have been designed and fast-response optical correlator has been created. The strong two-beam coupling ensured considerable amplification of weak coherent beams what was used for designing coherent optical oscillators. A very unusual oscillation dynamics have been discovered, with periodic π-shifts of the output wave phase what is a consequence of a pronounced bipolar conductivity of $Sn_2P_2S_6$. The bipolar conductivity manifests itself in particular spectral profile of two-beam coupling gain, with a narrow deep at exact frequency degeneracy. This allows also for slowing down of the short recording pulses (ms range) and speeding up of the long pulses (seconds and longer). The nanoparticles of $Sn_2P_2S_6$ suspended in liquid crystal produced a composite material with the enhanced optical nonlinearity. Finally, the efforts of French [2] and Chinese [3] research laboratories opened the way to successful use of $Sn_2P_2S_6$ for tumor cells imaging with combined ultrasound-optical testing technique.

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Surface Plasmons in Various Graphene Structures

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Graphene is well suited for a number of photonic applications due to its interesting optical and electronic properties. The intrinsic collective excitation in graphene has attracted much attention. Specifically, surface plasmons in graphene are believed to be potential ingredients for infrared and terahertz applications. Recently, graphene plasmons are not only theoretically investigated in structured graphene, such as graphene ribbons, coupled graphene ribbons, graphene ribbon arrays, graphene rings, and graphene disk arrays, but also studied experimentally in microribbon arrays and extended graphene sheets.

Different coupling effects including the mode coupling, phonon-photon coupling, and etc. are studied numerically for intentional purposes. The coupled modes between graphene plasmons and surface phonons of a semiconductor substrate are investigated, which can beneficently controlled by carrier injection of the substrate. The properties of surface plasmons localized at the interface between graphene and Kerr-type nonlinear substrates are investigated analytically. The dispersion of graphene plasmons may be affected much by the inevitable nonlinear effect of substrates. The spatial switching of mid-infrared light near-fields is proposed in coupled graphene heterogeneous ribbon pairs. By using the coupled plasmon modes in graphene ribbon pairs, the electric near-field enhancement can be spatially controlled in graphene ribbons as the tuning of the external bias voltage difference. Furthermore, plasmon induced transparency (PIT) effect is demonstrated in such graphene ribbon pairs. The transparency effect is understood by the mode coupling between dipolar and quadrupole plasmons modes in graphene ribbons. A typical Fano resonance for a metallic symmetry-breaking structure is simulated for graphene. Although a Fano-like extinction spectrum emerges, our analysis proves that the asymmetry is due to the intensity superposition of three plasmon modes instead of interference. The difference between graphene and metal plasmons comes from different contributions to the extinction, where the former is absorption instead of scattering.

Our research bases on graphene defects induced by ion beams, and near-field optical microscopy is employed in the investigation of plasmon propagation. Firstly, plasmon reflection near ion-induced graphene defect boundary was observed. Different degrees of defects were induced by varied ion doses, which results in tailorable properties of plasmon reflection. The realization of tailorable plamson reflection in this research opens up a new avenue for plasmon wave engineering. Secondly, graphene nanoresonators are fabricated easily and precisely with a spatial resolution better than 30 nm. Breathing modes are observed in graphene disks. The amorphous carbons around weaken the response of edge modes in the resonators, but meanwhile render the isolated resonators in-plane electrical connections, where near-fields are proved gate-tunable. The realization of gate-tunable near-fields of graphene 2D resonators opens up tunable near field couplings with matters. The realization of electrically tunable graphene nanoresonators by ion beam direct-writing is promising for active manipulation of emission and sensing at the nanoscale.

We proposed a new idea to improve the fabrication quality of graphene structures by the laser direct writing (LDW). Graphene slices grown by chemical vapor deposition (CVD) were transferred onto a SiC substrate. A Ti:sapphire femtosecond laser Gaussian beam (wavelength λ=800 nm, 120 fs pulse duration, 1 kHz repetition rate) was used to process the samples. The resolution could easily break the diffraction limit under proper ambient conditions. The width was reduced to as narrow as 216 nm and the precision was kept in ± 15 nm, only 11% of the resolution. Scattering scanning near-field optical microscope (s-SNOM) detecting was performed to verify that the fabrication forms a perfect reflection edge. The realization of the super-diffraction fabrication of graphene structures and the advantages of our method in near-field detection have a huge potential in structured graphene nano-photonics.

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Physical properties of multiferroic crystals under illumination

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A light-beam modifies the equilibrium tensors characterizing the physical properties of a crystal because it interacts with the crystal lattice. When considering the response to illumination at macroscopic time scales, i.e. much larger than the light period, only the static, or stationary, responses can be considered. Therefore, light represents an external field similar to the electric, magnetic or elastic applied fields, and the material appears to the light-beam as a continuum since the light wave length is much larger than the lattice spacing. Accordingly, the response of the crystal to illumination depends only on the point-group symmetries of the light-beam and of the crystal. In the last decades the influence of light on the properties of ferroic materials has essentially been focused on the *bulk photovoltaic effect* in ferroelectrics, and a number of physical properties have been highlighted such as domain-wall photovoltaics, above band gap photovoltage, or switchable diode-like effects.

In multiferroic materials the photovoltaic process is more complex as it involves the influence of light on the coupled magnetic and ferroelectric properties, and the reciprocal influence of these properties on the photovoltaic response. The current power expansion describing the bulk photovoltaic effect does not provide a direct method for combining the magnetic symmetry of the multiferroic crystal and the orientation and polarization of the light-beam wave. A more appropriate approach to the photovoltaic and other light-induced effects in multiferroics was recently proposed[1-3] based on an expansion of the relevant physical tensors in Wigner spherical functions. We will show that this approach allows predicting remarkable new effects in the paramagnetic and multiferroic phases of multiferroic crystals, namely *photo-magnetoelectric effects* corresponding to the emergence of light-induced components of the linear magnetoelectric tensor changing sign in opposite ferroelectric or magnetic domains, and *magneto-photovoltaic effects* characterized by photocurrents changing sense under space or time inversion. Light also induces changes in the elastic, electric and magnetic susceptibilities producing, for example, a *photoelastic* effect which consists of a deformation of the crystal under illumination.

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Optimizing Functional Nanostructured SolidsThrough Lattice Defect Engineering by Severe Plastic Deformation

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Over the last two decades the processing method “Severe Plastic Deformation–SPD” has impressively demonstrated that nanostructured materials with superior mechanical properties can be produced “top-down” in bulk shape which cannot be achieved with traditional “bottom-up” methods [1]. Now, the optimization of functional properties has been coming into the focus of the community’s research [2], not at least since outstanding successes such as world-records in the figure-of-merit (ZT) of SPD-thermoelectrics [3], and in the reproducibility in the hydrogen storage of SPD-processed hydrogen storage materials [4]. Recentinvestigations clearly suggest that a high density of SPD-induced lattice defects other than of classical grain boundaries can be equally or even more beneficial with respect to functional properties. For example, in case of thermoelectrics, SPD-induced dislocations and/or particular dislocation arrays seem to be most effective in increasing the ZT value [5]. Also in case of soft magnetic materials, regular dislocation arrays from SPD which form low-angle zero-strain nanocrystal boundaries promise new low-coercivity and high-magnetostriction materials [6,7], while in case of hydrogen storage, thermally stable SPD-induced vacancy clusters seem to govern the formation / dissolution of the hydride phase [8]. With the know-how to be obtained from systematic investigations, it should be possible to tailor specific defect structures on the nanoscale for optimum functional materials performances with very promising perspectives to practical application.

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Gamma Ray Spectroscopy at the Institut Laue-Langevin

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The ILL operates one of the most intense neutron sources in the world. Although primarily used for neutron scattering there exist a long-standing history of gamma ray spectroscopy at the ILL. The availability of well collimated intense neutron beams and in-pile sample irradiation positions allowed to develop quite unique instruments and applications of gamma ray spectroscopy. The talk will start with giving an overview on the different aspects of neutron based gamma ray spectroscopy at the ILL. Amongst the operating gamma ray instruments the ultra-high-resolution gamma ray spectrometers GAMS play a particular role due to their outstanding energy resolution and dynamic range. The instruments are based on perfect crystal Laue diffraction of gamma rays produced by excited nuclei in the reactor and their operation requires one of the world´s best angle measurement devices. Accordingly the experiments contributed in the past to many different fields in physics: nuclear structure, metrology, photon matter interaction, crystallography and astrophysics. In a second part the talk will review some highlights from the past 20 years of operation of these instruments.

$SrTiO_3$ – well known, but still surprising

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$SrTiO_3$ is intensively studied since more than 50 years. Different to the isostructural compound $BaTiO_3$ it does not become ferroelectric even at the lowest temperatures without applying an electric field. Its perovskite structure makes this compound particularly suitable as a substrate for high-$T_c$superconductors layers. The antiferrodistortive phase transition neat 105 K leads, however, to a domain structure with unusual properties: Due to higher order coupling effects, the domain distribution can not only be affected by mechanical stress but also by electric fields. In this presentation, it will be shown that both types of external load provide surprising effects which finally lead to the conclusion that even the electric field induced ferroelectric phase of $SrTiO_3$ is never a single domain phase. The combination of elastic and inelastic neutron scattering and Raman spectroscopy provides very detailed information about the microscopic behaviour. Moreover, the kinetics of the domain distribution under pulsed electric fields and mechanical stresses is studied by stroboscopic techniques.

Micro-structured liquid crystal alignment with out-of-plane polymer scaffolds

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Patterned alignment configurations for liquid crystals (LCs) are the core of many advanced LCD operation modes and of several non-display LC applications. Two-photon polymerization-based direct laser writing (TPP-DLW) is a very convenient technique to generate complex micro-structures for patterned LC alignment[1]. When introduced into the TPP-generated polymer scaffolds, the LC molecules are uniformly aligned via their contact to the walls of the polymer ribbons. The alignment is induced by surface relief gratings that are created on the polymer walls during the TPP-DLW process due to optical interference between the incident and reflected laser beams[2]. The associated surface anchoring energy is in the range of 10^{-5} J/m^2, which is considerably higher than observed on conventional surface relief grating materials.

With the aid of above mentioned polymer scaffolds, compartmentalized LC alignment in arbitrary 2D configurations can be realized. We present investigations of LC-based optical diffraction gratings exhibiting electro-optic and magneto-optic switching behavior (Fig.1) and electrically tunable q-plates for generation of vectorial vortex beams[3,4].

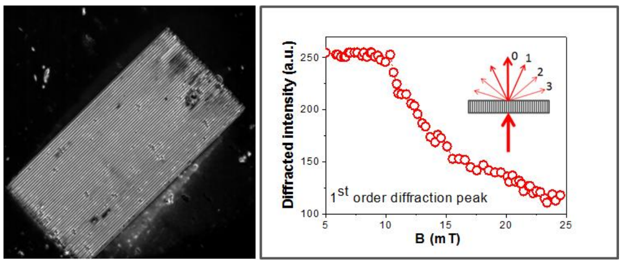

Fig. 1. Example of magnetically switchable liquid crystal grating structure.

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Tunable surface roughness and wettability of soft magnetoactive elastomers

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Dynamically responsive surfaces receive increasing attention in different areas of modern technology, such as touch-based interface systems and devices. Magneto-active elastomers (MAEs) belong to materials with promising properties for this group of applications [1,2]. MAEs are also suitable for some non-conventional applications, such as magnetically tunable substrates for biological cell cultures [3].

We investigated surface topographical modifications of a soft magnetoactive elastomer (MAE) in response to variable applied magnetic field. The analysis was performed *in-situ* and was based on optical microscopy, spread optical reflection and optical profilometry measurements. Optical profilometry analysis showed that the responsivity of magnetic field-induced surface roughness with respect to external magnetic field was in the range of 1 µm/T. A significant hysteresis of surface modifications took place for increasing and decreasing fields. Investigations of sessile water droplets deposited on the MAE surface revealed that field-induced topographical modifications affected the contact angle of water at the surface. This effect was reversible and its responsivity to magnetic field was in the range of 20°/T. Despite the increased surface roughness, the apparent contact angle decreased with increasing field, which we attributed to the field-induced protrusion of hydrophilic microparticles from the surface layer[4].

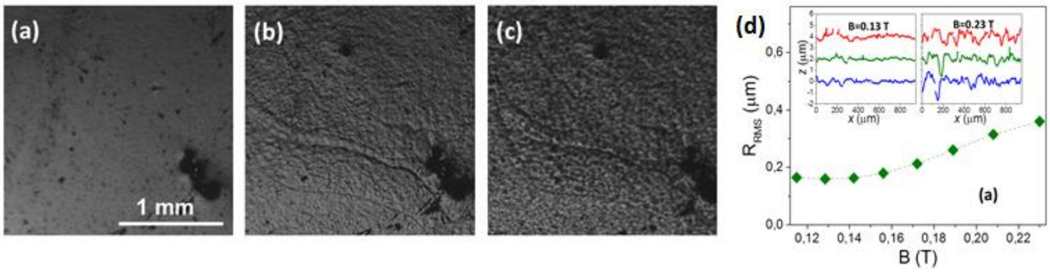

Fig. 1.Magnetic field-induced modifications of surface roughness.

This research has been partially supported by the Slovenian Research Agency (ARRS) and the German Academic Exchange Service (DAAD) bilateral project BI-DE/17-19-17, DAAD-ID 57364932; and by the joint research project between Slovenian Academy of Sciences and Arts and Hungarian Academy of Sciences ("Microfluidic systems based on anisotropic soft matter").

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Observation of multi-energy neutron matter-wave Pendellösung interference in holographic nanostructures

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Previous experiments with nanoparticle-polymer composite grating structures fabricated by optical holography have demonstrated how they can be used as multi-port beam-splitters or mirrors for cold and very-cold neutrons. In a recent experiment, we have observed the Pendellösung interference effect occurring simulataneously for the wavelengths of the broad spectrum of a very-cold (VCN) beam in the periodic potential of holographic nanostructures.

Dynamics of randomly pinned ferroelastic domain walls

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Dynamic Mechanical Analysis (DMA) measurements as a function of temperature, frequency and dynamic force amplitude are used to perform a detailed study of the domain wall motion in $LaAlO_3$. In previous DMA measurements Harrison, et al. [1] found evidence for dynamic phase transitions of ferroelastic domain walls in $LaAlO_3$. In the present work we focus on the creep-to-relaxation region of domain wall motion using two complementary methods. We determine, additionally to dynamic susceptibility data, waiting time distributions of strain jerks during slowly increasing stress. These strain jerks, which result from self-similar avalanches close to the depinning threshold, follow a power-law behaviour with an energy exponent $\varepsilon \approx 1.7$. Also, the distribution of waiting times between events follows a power-law with an exponent $n = 0.9$. The present dynamic susceptibility data can be well fitted [2] with a power law, with the same exponent ($n=0.9$) up to a characteristic frequency $\omega \sim \omega^*$, where a crossover from stochastic DW motion to the pinned regime is well described using the scaling function of A.A. Fedorenko, et al. [3].

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Investigations of the behaviour of supercooled water in nanopores and properties of polymers using Dynamic Mechanical Analizers

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Dynamic mechanical analyzers are very usefull for investigation of the dynamical properties of quite different materials in wide temperature region (-180°C to 600 °C) and frequency range 0.01 Hz - 100 Hz. DMA in most of all applications and measurement geometries tests Young's modulus Y , with stress and strain in the same direction. The force F is applied by a motor and transferred upon the samples surface via a metallic or quartz rod.Measurements can be performed in various geometries. For the molecular glass forming liquids and water we used mainly parallel plate (PP) and three-point bending (TPB) geometry and for themeasurements of polymers we mainly applied tension mode.

In this work we introduce the results of thermal expansion and low-frequency elastic measurements of polymers and supercooled water confined in nano-porous silica [1-5].

Acknowledgements: The present work was supported by the Austrian Science Fund (FWF, project Nr . P 28672 – N36).

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