

MO 7: Poster: Femtosecond Spectroscopy

Time: Tuesday 16:00–18:30

Location: Lichthof

MO 7.1 Tu 16:00 Lichthof

Vibrational wave packet dynamics of alkali molecules formed on helium nanodroplets — ●BARBARA GRÜNER¹, CHRISTIAN GIESE¹, LUTZ FECHNER¹, MATTHEW SIEVERT¹, MARTIN SCHLESINGER², MARCEL MUDRICH¹, and FRANK STIENKEMEIER¹ — ¹Physikalisches Institut, Universität Freiburg, Germany — ²Institut für Theoretische Physik, Technische Universität Dresden, Germany

Isolation in superfluid helium nanodroplets is a valuable tool for investigating molecules at temperatures in the millikelvin range as well as for studying decoherence mechanisms in weakly coupled molecule-bath systems. Using fs-pump-probe spectroscopy we study the dynamics of wave packets formed by the coherent superposition of vibrational states of both rubidium and potassium dimers and trimers. Wave packet dynamics is mass selectively detected after desorption off the helium nanodroplets and can be observed up to nanoseconds. Fourier analysis of the data reveals the vibrational modes of the high-spin alkali molecules (triplet states for dimers and quartet states for trimers), which are compared to ab initio calculations

MO 7.2 Tu 16:00 Lichthof

Femtosecond pulse shaping and quantum control in multi-channel molecular dynamics studied by coincidence imaging — ●CARL STEFAN LEHMANN, ARNO VREDENBORG, WIM G. ROETERDINK, and MAURICE H.M. JANSSEN — Laser Centre and Department of Chemistry, Vrije Universiteit, De Boelelaan 1083, 1081 HV Amsterdam, The Netherlands

The most complete information on molecular photodissociation and ionisation dynamics can be obtained by photo-electron / photo-ion coincidence imaging. Coincidence imaging is a technique in which both the ejected photo-electron and the ionic photofragment are measured, in coincidence, with two time- and position sensitive particle detectors. From the data the complete energetic and angular resolved distributions are obtained. In laser excitation of molecules with femtosecond pulses different multiphoton processes are easily induced leading to various ionic atomic and molecular fragment channels. The energy correlation of the electron with its correlated ionic fragment enables to distinguish the competing photon processes. We combine the coincidence imaging technique with the nowadays well-established pulse shaping technique. Pulse shaping provides a tool to control the reaction pathway, thereby controlling the reaction outcome. In this contribution the experimental set-up and the first experimental data on shaping the ultrafast induced dynamics in CH₂BrCl will be presented.

MO 7.3 Tu 16:00 Lichthof

Four-Wave Mixing Techniques with High Spatial Resolution — ●MAHESH NAMBOODIRI, VINU V NAMBOODIRI, GÜNTER FLACHENECKER, and ARNULF MATERNY — Jacobs University Bremen, Bremen, Germany

Coherent anti-Stokes Raman scattering microscopy (Micro-CARS) is a promising new technique for the imaging of specific molecular components, combining vibrational contrast of molecules with high spatial resolution [1]. This high resolution is due to the nonlinear interaction of three laser beams resulting in a cubic dependence of the signal on the laser intensity, which is highest in the laser focus. Due to the use of the microscope optic a collinear beam geometry has to be chosen, which is giving rise to an unwanted non resonant CARS background covering the signal. In our presentation, we show concepts and supported by experimental results suitable for a suppression of this background. Here, we mainly will focus on a femtosecond time-resolved three-color CARS. Additionally, we will discuss techniques for improving the spatial resolution of Micro-CARS.

References:

[1] A. Zumbusch, G.R. Holtom, X.S. Xie, *Phys. Rev. Lett.* **82**, 4142 (1999)

MO 7.4 Tu 16:00 Lichthof

Two-Photon Processes in Femtosecond Time-Resolved Four-Wave Mixing Spectroscopy: β -Carotene — ●VINU V NAMBOODIRI, MAHESH NAMBOODIRI, GÜNTER FLACHENECKER, and ARNULF MATERNY — Jacobs University Bremen, Bremen, Germany

Pump-degenerate four-wave mixing (pump-DFWM) using femtosecond pulses is used to study the dynamics of excited population (S_1)

and ground state vibrational relaxation of β -carotene molecules in different solvents. An initial pump (IP) pulse populates the S_2 state. The DFWM pulses are tuned to be in resonance with the $S_n \leftarrow S_1$ transition. Scanning the time delay between the IP and the DFWM sequence gives rise to a transient, which could not be explained up to now [1]. Recent experiments have shown that two-photon transitions have to be taken into account. A three-level kinetic model including a two-photon process is used to fit the transients and extract time constants. The time constants obtained are in agreement with previously reported values [2,3].

References:

[1] V. Namboodiri, A. Scaria, M. Namboodiri, A. Materny, *Las. Phys.* **19**, 154 (2009).

[2] J. Hauer, T. Buckup, M. Motzkus, *J. Phys. Chem. A* **111**, 10517 (2007).

[3] D. W. McCamant, J.E. Kim, R.A. Mathies, *J. Phys. Chem. A* **106**, 6030 (2002).

MO 7.5 Tu 16:00 Lichthof

Detailed simulation of time-resolved four wave mixing in carotenoids using the Brownian oscillator model — ●MARIE MAREK^{1,2}, TIAGO BUCKUP^{1,2}, and MARCUS MOTZKUS^{1,2} — ¹Physikalisch-Chemisches Institut, Ruprecht-Karls-Universität Heidelberg, D-69120 Heidelberg, Germany — ²Physikalische Chemie, Philipps-Universität, D-35043 Marburg, Germany

Degenerate four wave mixing (DFWM) is an ideal technique for the simultaneous investigation of population and vibrational dynamics in complex molecular systems. However, for the interpretation of the four wave mixing signal a firm theoretical support is crucial due to the various processes contributing to the signal generation. In this work we develop systematically a fitting and simulation tool for DFWM based on the Brownian oscillator model. Depending on the underlying model and the investigated system up to 80 different interactions can be considered. We demonstrate our approach by fitting the signal of several carotenoids using an evolutionary algorithm, which allows for the testing of several rate models describing the carotenoids relaxation pathways. The best agreement is obtained with a simple model containing only S_2 , S_1 and S_0 with the respective lifetimes known from literature. Furthermore, our fitting procedure permits to determine the ratio of the involved transition dipole moments. Finally, we extend the simulations to pump-DFWM by introducing a population shift generated by the initial pump pulse and then calculating the DFWM signal on the excited state. By this means we confirm the involvement of a low-lying vibrationally hot state in the deactivation process of carotenoids.

MO 7.6 Tu 16:00 Lichthof

Pump-probe experiments on H_2 ultrashort carrier-envelope phase stabilized laser pulses — ●VANDANA SHARMA¹, BETTINA FISCHER¹, MANUEL KREMER¹, BERNOLD FEUERSTEIN¹, THOMAS PFEIFER¹, UWE THUMM², ROBERT MOSHAMMER¹, and JOACHIM ULLRICH¹ — ¹Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany — ²James R. Macdonald Laboratory, Kansas State University, Manhattan, Kansas 66506-2606, USA

The ability to control electron localization in H_2 dissociating due to bond-softening by carrier-envelope phase stabilized ultrashort laser pulses was recently demonstrated experimentally [1]. In order to gain more control and more insight into the localization mechanism, the measurements were extended to two laser pulses. By the first pulse the neutral H_2 molecule is singly ionized and a wave packet is created in H_2^+ . Electron localization is achieved via a second laser pulse at a variable time delay after the first one. Studying the delay-dependent electron localization in the revival of the wave packet allows one to gain insight into the influence of the wave packet dynamics. By a comparison of the revival with the dephasing region the influence of coherence is analyzed. Measurements on the delay-dependent proton emission asymmetry in the revival and the dephasing region are presented. Comparing the data to wave packet simulations allows to attribute the structure of the asymmetry to the properties of the wave packet.

[1] Manuel Kremer et al., *Phys. Rev. Lett.* **103**, 213003 (2009)

MO 7.7 Tu 16:00 Lichthof

Transient absorption spectroscopy of a metallo-supramolecular polyelectrolyte — ●TATJANA QUAST¹, FLORIAN KANAL¹, IRIS HASSLAUER², DIRK G. KURTH², and TOBIAS BRIXNER¹ — ¹Institut für Physikalische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg — ²University of Wuerzburg, Chemical Technology of Advanced Materials, D-97070 Würzburg

Supramolecular chemistry is becoming increasingly important for the design and development of new functional materials. With the help of self-organization, one can create stimuli-responsive materials, which change their properties upon an external stimulus (e.g. change of pH value, temperature). By including metal ions, one can modify the magnetic, optical and electronic properties of these materials. Here we present a metallo-supramolecular polyelectrolyte (MEPE), which is constituted of a ditopic Bisterpyridin ligand and a transition metal ion, in this case Fe. This Fe-MEPE forms rigid, rod-like polymers and exhibits a strong metal-to-ligand charge transfer (MLCT) band in the visible spectral range. In order to monitor the dynamics of Fe-MEPE after excitation, we recorded transient absorption spectra of Fe-MEPE in aqueous solution. We excited the MLCT band of the system with a laser pulse from an optical parametric amplifier and probed the dynamics of this band with a whitelight supercontinuum. First results on the dynamics of the Fe-MEPE polymer and the corresponding monomer are presented.

MO 7.8 Tu 16:00 Lichthof

Fragmentation studies of chiral molecules via femtosecond-laser mass spectrometry — ●CHRISTIAN LUX, QINGQING LIANG, CRISTIAN SARPE-TUDORAN, MATTHIAS WOLLENHAUPT, and THOMAS BAUMERT — Universität Kassel, Institut für Physik und CINSaT, D-34132 Kassel, Germany

Under symmetrical conditions two enantiomers of a chiral molecule that are non superimposable mirror images of each other show almost the same physical properties. In the recent past the analysis of enantiomers based on laser irradiation using circular polarized femtosecond-laser pulses was realized for many chiral molecules. Circular dichroism (CD) refers to the difference between absorption of left- and right-circularly polarized light for one enantiomer. Absorption values for both enantiomers show a specific anisotropy. In this contribution we study the effect of CD on the ion yield of chiral molecules by mass spectrometry via ultrashort laser pulses [1-3]. We used a home-build Wiley-McLaren time-of-flight mass spectrometer and infrared circularly-polarized femtosecond-laser pulses to measure the intensity dependence of mass-spectra from low-volatile chiral substances in particular for the enantiomers of camphor.

- [1] U. Boesl *et al.*, Chem. Phys. Chem. 7: 2085-2087 (2006)
 [2] A. Bornschlegel *et al.*, Chem. Phys. Lett. 447: 187-191 (2007)
 [3] H. G. Breuning *et al.*, Chem. Phys. Chem. 10: 1199-1202 (2009)

MO 7.9 Tu 16:00 Lichthof

Comparison of wave-packet dynamics in Bacteriorhodopsin and all-trans Retinal Protonated Schiff-Bases using Degenerate Four-Wave-Mixing — ●JAN PHILIP KRAACK^{1,2}, TIAGO BUCKUP¹, and MARCUS MOTZKUS^{1,2} — ¹Physikalisch-Chemisches Institut, Ruprecht-Karls-Universität Heidelberg, D-69120 Heidelberg, Germany — ²Physikalische Chemie, Philipps-Universität, D-35043 Marburg, Germany

A prominent example for a light-harvesting system that converts light into chemical energy is Bacteriorhodopsin (bR) containing the all-trans retinal chromophore. Photon-absorption triggers a sub-picosecond, position-selective isomerization in protein-environment with a high quantum yield (0.65). This is contrasted by a slower (several picoseconds), unselective and inefficient isomerization of isolated all-trans retinal protonated Schiff-base (RPSB) in solution. This selectivity enhancement and acceleration effect of the protein environment has been a question of interest for a long time. We studied population and vibrational coherence dynamics on the sub-20 fs timescale using Degenerate Four-Wave-Mixing (DFWM) in order to clarify differences and analogies in the respective relaxation dynamics. We present detailed investigations on ground-state vibrational coherence dynamics and discuss the origin of excited state wave packet dynamics. Due to initial relaxation (<300 fs) and wave packet dynamics, we discuss the applicability of the 2-State-Two-Mode-Model [1], generally assumed to describe the relaxation dynamics of retinal-chromophores.

- [1] González-Luque *et al.*, PNAS 97, 2000, 9379

MO 7.10 Tu 16:00 Lichthof

Observation of vibrational wave packet dynamics in MgH⁺ —

●STEFFEN KAHRA¹, GÜNTHER LESCHHORN¹, HSIANG-TAI DOU¹, TOBIAS SCHAEZT¹, AGUSTIN SCHIFFRIN², RALPH ERNSTORFER², REINHARD KIENBERGER², MARKUS KOWALEWSKI³, and REGINA DE VIVIER-RIEDLE³ — ¹Max-Planck-Institut für Quantenoptik, Garching, Tlamo — ²MPQ, Garching, AS beam lines — ³Department Chemie and Biochemie, LMU München

We report measurements that demonstrate the combination of precise spatial control and initialization of single molecular ions in a Paul trap with temporal resolution provided by <5 fs UV laser pulses. In this proof of method experiment we show how the temporal evolution of a vibrational wave packet (vibrational period 31 fs) in a few distinguishable isolated molecules can be followed by a dissociative pump probe scheme. We describe the way to load and identify individual molecular ions in our trap and especially concentrate on how to count them reliably before and after the pump probe delay dependent dissociation. It is this time dependence that reflects the molecular motion and gives rise to our experimental signal. Additionally to the vibrational content of the measured oscillation curve recent theoretic insight predicts that fast components from electronic relaxation processes will contribute significantly to the signal. A similar experiment using a different excitation spectrum in the UV might allow us to distinguish between the different contributions to the oscillatory signal and thus spotlight the fast dynamic components.

MO 7.11 Tu 16:00 Lichthof

Suppression of perturbed free induction decay and noise in experimental ultrafast pump-probe data — ●PATRICK NUERNBERGER, KEVIN F. LEE, ADELIN BONVALET, THOMAS POLACK, MARTEN H. VOS, ANTIGONI ALEXANDROU, and MANUEL JOFFRE — Laboratoire d'Optique et Biosciences, Ecole Polytechnique, Centre National de la Recherche Scientifique, 91128 Palaiseau, France, and Institut National de la Santé et de la Recherche Médicale, U696, 91128 Palaiseau, France

We apply a Fourier-filtering technique for the global removal of coherent contributions, like perturbed free induction decay, and noise, to experimental pump-probe spectra. A further filtering scheme gains access to spectra otherwise only recordable by scanning the probe's center frequency, with adjustable spectral resolution. These methods cleanse pump-probe data and allow improved visualization and simpler analysis of the contained dynamics. We demonstrate these filters using visible pump/midinfrared probe spectroscopy of ligand dissociation in carboxy-hemoglobin.

MO 7.12 Tu 16:00 Lichthof

Two-dimensional spectroscopy of individual single-wall carbon nanotubes — ●ULRIKE SELIG, NADJA BERTLEFF, DOMINIK STICH, TOBIAS HERTEL, and TOBIAS BRIXNER — Institut für Physikalische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg

Possible applications as nano-electronic devices or in new composite materials have caused considerable interest in carbon nanotubes and their unique electronic and mechanical properties [1,2]. One of the outstanding features of individual single-wall carbon nanotubes (SWNT) is their quasi one-dimensional (1D) band-structure.

Here we present first results of degenerate pump-probe experiments and two-dimensional spectra of (6,5) SWNTs excited with 30fs pulses with a center wavelength of 565nm, along with an interpretation of the observed relaxation processes from the S_2 electronic state.

- [1] R. Saito, G. Dresselhaus, M. Dresselhaus, Physical Properties of Carbon Nanotubes, London, Imperial College Press, 1998.

- [2] T.W. Ebbesen, Carbon Nanotubes, Preparation and Properties, CRC Press, Boca Raton, FL, 1997.

MO 7.13 Tu 16:00 Lichthof

Challenges in single-particle diffractive imaging with intense Xray pulses — ●MARTIN WINTER, CHRISTIAN GNODTKE, ULF SAALMANN, and JAN-MICHAEL ROST — Max Planck Institut fuer Physik komplexer Systeme, 01187 Dresden, Germany

The construction of short-wavelength free-electron lasers (FEL) allows for the investigation of intense laser-matter interaction at the so far unexplored Xray regime. The FEL at LCLS/Stanford is the first one reaching short intense pulses with photon energy in the keV range. One of the main objectives of these machines is the imaging of single molecules by measuring diffraction images from exposure by the beam.

We will discuss two complementary issues which have to be overcome to obtain diffraction images whereof the molecular structure can

be successfully reconstructed. 1) The damage, in particular the ionization of the sample, induces an explosion on the time scale of the pulse. We discuss the impact of a tamper [1,2] which slows down the expansion and thus may improve the diffraction image. 2) Due to the small elastic-scattering cross sections diffraction images obtained will be very faint, which makes the determination of the orientation of the molecule challenging. We have implemented a method based on the generative topographic mapping [3] and will discuss its application to two-dimensional diffraction images.

References: [1] C. Gnodtke, U. Saalman and J. M. Rost, Phys. Rev. A 79, 041201 (2009) [2] S. P. Hau-Riege et al., Phys. Rev. Lett. 98, 198302 (2007) [3] R. Fung, V. Shneerson, D. K. Saldin and A. Ourmazd, NPHYS 1129 (2008)

MO 7.14 Tu 16:00 Lichthof

Elektronwinkelverteilung bei Ionisationsprozessen von H_2^+ in starken Laserfeldern — MATTHIAS ODENWELLER, LOTHAR PH. H. SCHMIDT, KYRA COLE, ●JASMIN TITZE, ARNO VREDENBORG, HONG-KEUN KIM, JÖRG VOIGTSBERGER, KEVIN PAHL, HENDRIK SANN, TOBIAS BAUER, MORITZ MECKEL, TILO HAVERMEIER, NADINE NEUMANN, FELIX STURM, DANIEL METZ, ANNIKA JUNG, HORST SCHMIDT-BÖCKING und REINHARD DÖRNER — Institut für Kernphysik, Goethe Universität, Frankfurt am Main, Deutschland

H_2^+ stellt mit zwei Protonen und einem einzigen Elektron das einfachste Molekül dar. Trotzdem führten experimentelle Herausforderungen bisher dazu, dass es nur äußerst wenige Untersuchungen der Elektronendynamik gab.

In Reaktionen mit 40 fs Laserpulsen, der Intensität $7 \cdot 10^{14} \frac{W}{cm^2}$ konnten nun die Elektronwinkelverteilung bezüglich der molekularen Achse und der Polarisation gemessen werden. Ein speziell für dieses Experiment entwickeltes Spektrometer erlaubt die effiziente Trennung von Target-Elektronen und Elektronen, die vom Laser aus dem Restgas erzeugt werden. In das Spektrometer ist des Weiteren ein Monitorsystem für die Position des Laserfokuses sowie für die Strahlage des Ionenstrahls integriert. Dank einer an die renormierte COLTRIMS-Technik (COLd Target Recoil Ion Momentum Spectroscopy) angelehnten Messmethode konnten die Impulse aller beteiligten Teilchen dabei über den gesamten Raumwinkel gemessen werden.

MO 7.15 Tu 16:00 Lichthof

Sub-Picosecond Time-Resolved Infrared Spectroscopy of 3-Hydroxyflavone in solution — ●KATHARINA CHEVALIER, MATTHIAS M.N. WOLF, and ROLF DILLER — Fachbereich Physik, TU Kaiserslautern, D-67663 Kaiserslautern

The properties of 3-hydroxyflavone and its intramolecular proton transfer after electronic excitation have been subject to numerous investigations, including stationary FTIR, femtosecond fluorescence and transient absorption spectroscopy in the visible.

Here we present to our knowledge the first results of sub-picosecond time-resolved IR vibrational spectroscopy of the photoinduced processes in 3-hydroxyflavone in solution. The results yield direct insight into photoinduced dynamics and structural information on the respective educt and product states. The observed vibrational bands can be compared to results from infrared/resonant 2-photon ionisation (IR/R2PI) experiments in gas phase [1].

[1] K. Bartl, A. Funk, K. Schwing, H. Fricke, G. Kock, H.-D. Martin, M. Gerhards, Phys. Chem. Chem. Phys. 2009, 11, 1173

MO 7.16 Tu 16:00 Lichthof

CRASY: Correlated Rotational Alignment Spectroscopy Resolves Fragmentation Channels — ●CHRISTIAN SCHRÖTER, KYRIAKI KOSMA, INGOLF-VOLKER HERTEL, and THOMAS SCHULTZ — Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin

Mass spectroscopy yields information on the composition of molecules and clusters. When the formed compounds are weakly bound they may break apart which leads to an increased ion yield in the fragment. Here we address the question how to distinguish between fragmented and unfragmented signals.

We developed a CRASY method which combines the techniques of rotational alignment with femtosecond-pump-probe spectroscopy in a two pulse experiment. An IR pulse generates a coherent rotational wave packet by means of nonadiabatic alignment. After a variable delay, we probe the wave packet by a UV pulse which excites and ionizes the molecule via a resonant electronic state. Every ion signal is modulated with a rotational frequency linked to the ground state geometry,

since the transition dipole moments are fixed in the rotating molecular frame. This mass-CRASY method yields mass-rotational spectra.

We demonstrate the technique using CS_2 as a model system with distinct fragmentation channels and discuss fragmentation paths for selected rotational states.

MO 7.17 Tu 16:00 Lichthof

Electronic Structure and Dynamics of Cytosine — KYRIAKI KOSMA, ●CHRISTIAN SCHRÖTER, INGOLF-VOLKER HERTEL, and THOMAS SCHULTZ — Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin

The characterization of the relaxation processes in isolated DNA bases (adenine, thymine, cytosine, and guanine) may help to understand the photostability of DNA upon UV irradiation.

For cytosine three tautomers have been observed in the gas phase: the enol, keto, and keto-imino forms [1]. We studied the electronic structure and dynamics of these tautomers by means of time-resolved mass and photoelectron spectroscopy.

Three ionic transients with lifetimes of femtoseconds to hundreds of picoseconds lifetimes are observed for the biologically relevant keto tautomer and are correlated with broad, unstructured electron bands. The data are assigned to internal conversion and excited-state tautomerization or intersystem crossing. Only two transients with femtosecond and picosecond lifetimes are identified for the enol or keto-imino tautomer and are assigned to internal conversion processes [2].

[1] Brown R. D. et al., J. Am. Chem. Soc. 1989, 111, 2308

[2] Kosma K. et al, J. Am. Chem. Soc. 2009, 131, 16939

MO 7.18 Tu 16:00 Lichthof

Dynamics of elementary excitations in para-hydrogen crystals: long living, coherent phonons, rotons and stimulated rotational Raman beats — ●FALK KÖNIGSMANN, NINA OWSCHIMIKOW, and NIKOLAUS SCHWENTNER — Freie Universität Berlin, Institut für Experimentalphysik, Arnimallee 14, D-14195 Berlin

One of the most distinct spectroscopic features of cryogenic para-hydrogen are the sharp linewidths. The thus expected very long coherence times in the dynamics did however not show up in previous ultrashort pulsed excitations. Our approach is ultrashort, spectrally resolved Optical Kerr Effect (OKE) spectroscopy, sensitive to induced anisotropies. We grow 3 cm long, transparent para-hydrogen crystals in an enclosed cell at 10 K, which are then cooled down to 4 K. The crystals are pumped with 150 fs pulses at 780 nm of a Ti:Sa amplified laser system and the induced birefringence is detected with the second harmonic in a colinear way. We observe a long lasting (>20 ps) sinusoidal birefringence modulation with a period of 907 fs. It coincides with the transverse, optical phonon at the center of the Brillouin-zone, which is observed exclusively in Raman scattering. We also observed long lasting (>80 ps), higher frequency birefringence modulations with a period of 94 fs and pump-induced, stimulated Stokes- and Anti-Stokes Raman bands of the detected second harmonic. Both match the J=2 roton excitation of 354 wavenumbers. The latter carries a 17 ps beating structure, which corresponds to the 2.0 wavenumbers splitting in the roton triplet also known from cw Raman-spectroscopy.

MO 7.19 Tu 16:00 Lichthof

Rotational decoherence in nitrogen investigated by non-adiabatic alignment — ●ANNA OTT, NINA OWSCHIMIKOW, FALK KÖNIGSMANN, and NIKOLAUS SCHWENTNER — Institut für Experimentalphysik, Freie Universität Berlin

The superposition of rotational states in a wave packet upon excitation of a molecule with anisotropic polarizability with a short, intense laser pulse leads to the appearance of periodic revivals of alignment. We detect alignment in nitrogen gas by the ultrafast Kerr effect method and observe the decay of the coherent signal induced by collisions in pure nitrogen and in gas mixtures of nitrogen with hydrogen and argon. From the decay rates we extract cross sections for decoherence, containing the sum of the effects of energy transfer and pure dephasing. The cross sections decrease with increasing temperature, leading to long lived coherence at high temperatures. Quantitative comparison with depopulation cross sections from measurements of linewidth broadening of the Raman Q-Branch in the literature shows that decoherence in rotational alignment decays at the same rate as the excited population. Thus, in the investigated system there occurs no pure rotational dephasing and all decoherence events are connected with the transfer of energy.