

# Femtosecond spectroscopy of the $(2) \ ^1\Sigma_u^+$ double minimum state of $\text{Na}_2$ : time domain and frequency spectroscopy

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**Abstract.** Femtosecond time resolved pump-probe experiments studying wave packet dynamics in the  $(2) \ ^1\Sigma_u^+$  double minimum state of  $\text{Na}_2$  are reported. The experiments were performed in a molecular beam with ion Time of Flight (TOF) detection. By Fast Fourier Transformation (FFT) of the observed time domain data the energy spacings of the coherently coupled vibrational levels in the  $(2) \ ^1\Sigma_u^+$  potential are obtained with an accuracy of  $0.02 \text{ cm}^{-1}$ , although an ultrafast laser source with its inherent spectral width was used in the experiment.

The wavelengths of the pump and probe laser pulses are chosen such that in this two color experiment we can control ionisation versus ionisation induced fragmentation.

In order to study the influence of the potential barrier on a vibrational wave packet motion we performed simulations based on time dependent quantum calculations.

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## 1 Introduction

By coherent coupling of vibrational states of a molecule a vibrational wave packet is formed. The evolution of the phases of these vibrational eigenfunctions leads to a classical oscillatory motion of the wave packet. Erwin Schrödinger already realized this [1] shortly after he had published his Schrödinger equation. In this publication he formed a wave packet in a harmonic oscillator potential and showed this wave packet moves like a point mass according to the laws of classical mechanics.

With the development of ultrafast laser sources with their inherent spectral width it has become a standard technique to coherently couple quantum mechanical eigenstates. The observation of Rydberg wave packets in atoms [2, 3], showing the classical Kepler orbits of the electron around the nucleus [4], and the observation of vibrational wave packets in molecules [5–10] are prominent examples.

In this contribution we discuss several aspects of vibrational wave packet motion. As an example we have chosen the physically attractive case of the  $(2) \ ^1\Sigma_u^+$  double minimum potential well in the  $\text{Na}_2$  molecule. In future experiments we will study the influence of the potential barrier on wave packet motion. Basic quantum optical effects such as the splitting of a wave packet at the barrier are expected. Until now we have studied the wave packet dynamics above the barrier and we will discuss our results from two different point of views. First we will describe how frequency spectroscopy can be performed in the time domain in spite of the inherent spectral width of an ultrashort laser pulse. A bandwidth limited 50 fs Gaussian laser pulse, for example, inhibits a spectral width of about  $300 \text{ cm}^{-1}$ . Secondly we will discuss the classical aspects of vibrational wave packet motion, because femtosecond pump-probe techniques provide an ideal means for the investigation and control of molecular motion in real time [11]. The internuclear distance in a diatomic molecule can be controlled by adjusting the delay time of pump and probe laser pulses. Probe transitions at small internuclear distances and at large distances lead to two different product channels (ionisation versus ionisation induced fragmentation).

At first sight the terms frequency spectroscopy and time domain measurements seem to be contradictory because of the broad spectral distribution of an ultrashort laser pulse. However, spectroscopic information can be derived by Fourier transformation of data taken in the time domain. This has been shown for diatomics and diatomic like molecules by Zewail's group for the systems  $\text{I}_2$  [12] and  $\text{ICN}$  [13]. In our group we have applied this technique to the high lying  $(4) \ ^1\Sigma_g^+$  shelf state in  $\text{Na}_2$  [14]. In addition we have extended this concept for the first time to triatomics. On the nonlinear  $\text{Na}_3$  molecule we were able to derive the normal modes of the X and B state [15] by Fourier transformation of the observed time domain data. As we will show in this contribution, by using ultrafast laser techniques the time domain approach can in principle compete with the elaborate techniques of high resolution spectroscopy even for bound systems. But until

now the methods of high resolution spectroscopy are far more developed and cheaper in comparison to time resolved laser methods. For predissociating or dissociative systems however, the time resolved approach may sometimes be the only choice in order to determine spectroscopic data especially in the transition state region. Another advantage of time resolved experiments, particularly in the case of congested spectra, is that it is easier to distinguish between vibrational and rotational spectroscopic information, because the energy spacings (e.g. oscillation periods) generally differ by two orders of magnitude. Additionally the time resolved method is intrinsically Doppler free. For bound systems the ultimate resolution is only limited by the scan length and the natural linewidth. Using a square window in the Fourier transformation, the theoretical resolution limit (FWHM) is  $0.1 \text{ cm}^{-1}$  for a scan length of 300 ps. Note that peak positions (frequencies) in such a FFT spectrum can be determined with even higher accuracy.

Controlling a chemical reaction such that a given product is produced at the expense of another, energetically allowed product is one of the basic issues in physical chemistry [16, 17]. For small molecular systems the basic ideas of the Tannor-Kosloff-Rice scheme [18] are applicable. These authors have proposed that controlling the duration of propagation of a wave packet on an excited electronic potential energy surface -i.e. controlling the internuclear distance- by simply controlling the time delay between pump and probe pulses, can be used to generate different chemical products. This idea was already confirmed by Zewail and coworkers [19]. In their experiment two sequential coherent laser pulses were used to control the reaction of  $\text{I}_2$  molecules with Xe atoms to form  $\text{XeI}$ . In the course of *one color femtosecond pump-probe* experiments on the multiphoton ionisation and fragmentation of  $\text{Na}_2$  [7] [20] we were able to demonstrate the control of the internuclear distance of the molecule. This control led to a variation in the ratio of dimer ions versus atomic fragment ions, showing that the Tannor-Kosloff-Rice scheme is valid [21, 22]. In this contribution we present a *two color femtosecond pump-probe* set-up used in the control experiments.

Quantum optical phenomena of wave packet motion are best investigated in molecular potentials, since the radiative lifetime of excited molecular states is of the order of nanoseconds and therefore long compared to typical vibrational oscillation periods which are of the order of several hundred femtoseconds. The long time behaviour of vibrational wave packet motion such as the spreading and recurrence of a vibrational wave packet has already been studied in our group experimentally [23]. To study the influence of a potential barrier on the vibrational wave packet motion is one of our next experimental goals. Quantum mechanical simulations are extremely helpful in understanding the control schemes and the quantum optical properties of wave packets in detail. To describe the propagation of vibrational wave packets in time we use the so called split-operator technique described by Kosloff [24] and Engel [25]. In this contribution we simulate the influence of the barrier on the wave packet motion in the  $(2) ^1\Sigma_u^+$  state of  $\text{Na}_2$ .

## 2 Experiment

A detailed description of our experimental set-up can be found in [26]. In the following we briefly describe the set-up used in the experiment.

In our femtosecond laser-molecular beam studies of time-resolved multiphoton ionisation and fragmentation processes, we use a combination of different experimental techniques. Femtosecond pump-probe techniques are used to induce and to probe molecular transitions, to resolve the interactions, to display the evolution of coherences and populations in real-time and to derive spectroscopic data from time domain measurements. A supersonic beam generates the molecules in the very lowest vibrational and rotational states. Time of Flight (TOF) spectroscopy is used to analyse the final continuum states by measuring the released kinetic energy of the ionic fragments and the energy distribution of the ejected electrons. The supersonic beam is produced either by a pure sodium expansion through a small orifice of typically 0.1 mm diameter or by an expansion seeded with argon. The oven is operated up to 1000 K with nozzle temperatures about 50 K higher. This technique provides efficiently cooled sodium molecules. Typical measured initial conditions were a population of 90% in  $v'' = 0$ .

Independently tunable femtosecond pulses down to 50 fs time duration and up to 50  $\mu\text{J}$  energy are generated in the home built laser system shown in Fig. 1. The tunability of this system covers the near UV, the complete visible range and the near IR. The output pulses of a colliding-pulse mode-locked ring dye laser (CPM) are amplified in a bow-tie amplifier which is pumped by an excimer laser at 308 nm, then pulse compressed and focused into a cell containing methanol to generate a white light continuum. Pump and probe pulses at specific wavelengths are selected from the white light continuum in a grating arrangement [27], which can also be used to compensate for group velocity dispersion in the subsequent amplification stages. By using adjustable slits for wavelength selection, the bandwidth of the pulses can be chosen. Pump and probe pulses are amplified again in two additional bow tie amplifiers. The pump laser beam at a wavelength of 680 nm is amplified before the frequency doubling. The resulting laser pulses at 340 nm have a spectral width of 4 nm FWHM (corresponding to  $350 \text{ cm}^{-1}$ ). The 540 nm (6.5 nm FWHM) probe laser pulses are also amplified. Pump and probe laser beams are recombined collinearly and focused into the interaction region. The laser beams, the molecular beam and the TOF spectrometer axis are mutually perpendicular in this set up. A Michelson arrangement is used to delay the probe laser pulse relative to the pump laser pulse. The achieved phase sensitive time resolution is about 1 fs as measured by interferometric autocorrelations [14].

## 3 Results and discussion

Before we start to discuss the topics of this contribution we first summarize the spectroscopy of the  $(2) ^1\Sigma_u^+$  double minimum state and describe the excitation scheme of the experiment.

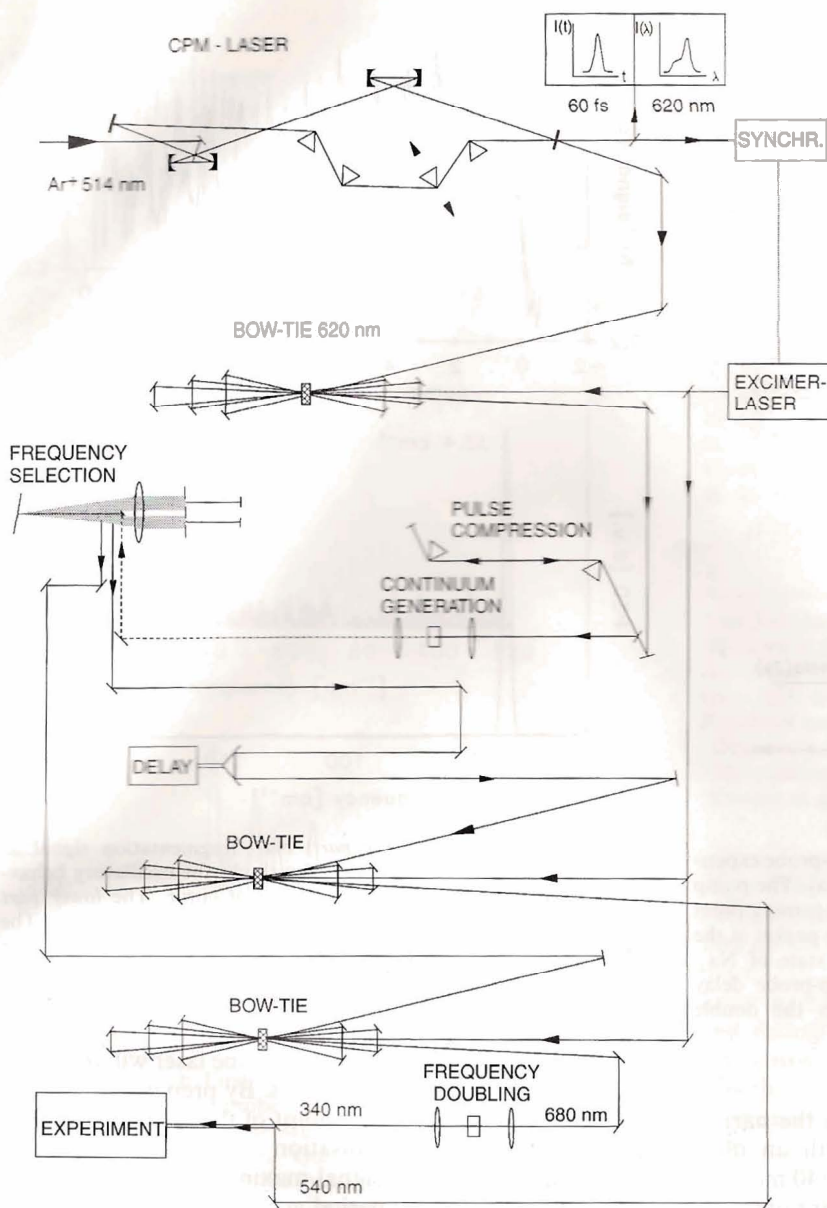


Fig. 1. Femtosecond laser system for independently tunable pump (340 nm) and probe (540 nm) wavelengths

The double minimum structure of the  $(2) \ ^1\Sigma_u^+$  state of  $\text{Na}_2$  (Fig. 2) is formed by the avoided crossing of two adiabatic potential curves. One has mainly Rydberg character and the other has substantial ionic character at large internuclear distances. The  $(2) \ ^1\Sigma_u^+$  state was theoretically predicted by Valance and Nguyen Tuan [28] and confirmed by Jeung [29]. The first experimental observation was reported by Cooper et al. [30] and Vergès et al. [31]. They employed Fourier transform spectroscopy of the laser induced infrared fluorescence and observed five vibrational levels of the inner well, all of the 29 levels of the outer well and some 65 upper levels. These observed levels were used to construct an adiabatic potential leading to the following molecular constants: the inner potential minimum is at an internuclear distance of 3.688 Å and at an energy of 28454.56  $\text{cm}^{-1}$  ( $\omega_v = 106.02 \text{ cm}^{-1}$ ), the outer potential minimum is located at 6.739 Å with an energy of 27879.40  $\text{cm}^{-1}$  ( $\omega_e = 52.60 \text{ cm}^{-1}$ ). The barrier

is at 4.716 Å with an energy of 29132  $\text{cm}^{-1}$ . The top of the barrier corresponds to an excitation wavelength of 344.2 nm relative to the  $X \ ^1\Sigma_g^+(v'' = 0)$  state.

Experiments involving the two photon ionisation (TPI) technique were performed first by Delacretaz and Wöste [32]. They observed all levels of the inner well and the 25 first levels above the potential barrier. The analysis of the  $\text{Na}^+$  signal in that work indicated significant fragmentation during the ionisation step for those  $\text{Na}_2$  molecules which were excited to levels above the potential barrier. Above the barrier the vibrational structure of the nearby  $C \ ^1\Pi_u$  state was seen in the recorded  $\text{Na}_2^+$  spectra, whereas in the recorded  $\text{Na}^+$  signal this structure was not dominant. In order to explain these findings the authors favoured a further excitation of the  $(2) \ ^1\Sigma_u^+$  state at large internuclear distances into the  $2^2\Sigma_u^+$  of  $\text{Na}_2^+$ . Later TPI experiments of Haugstätter et al. [33] confirmed this explanation.

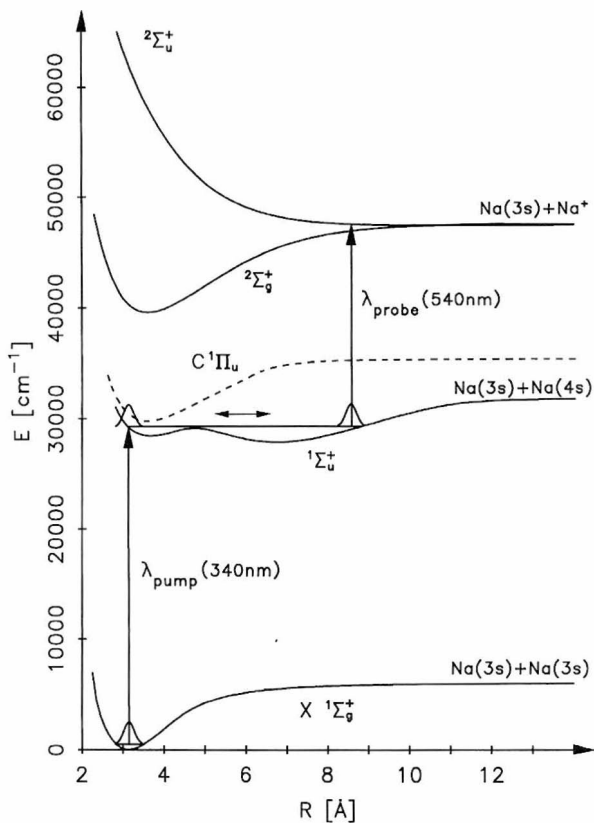


Fig. 2. Potential energy curves relevant for the pump-probe experiment on the  $(2) \ ^1\Sigma_u^+$  double minimum state of the  $\text{Na}_2$ . The pump laser prepares a vibrational wave packet at the inner turning point above the barrier. The probe laser transfers this wave packet at the outer turning point onto the repulsive  $2\Sigma_u^+$  ionic state of  $\text{Na}_2$ . Measuring the  $\text{Na}^+$  signal as a function of pump-probe delay therefore resembles the wave packet dynamics in the double minimum state

In order to prepare a wave packet experimentally, we excited the molecule with an ultra short pump pulse at a central wavelength of 340 nm. The wave packet is created at the inner turning point of the  $(2) \ ^1\Sigma_u^+$  state, as we start from  $v'' = 0$  in the narrow  $X \ ^1\Sigma_g^+$  state (Fig. 2). Starting from higher  $v''$  levels is unlikely since we have measured 90% of the population to be in  $v'' = 0$ . The calculated Franck-Condon Factors for the  $X \ (2) \ ^1\Sigma_u^+$  transition, using the potential derived by Cooper et al. [30], support also that  $v'' = 0$  is the initial vibrational level for the excitation. The Franck Condon maximum for  $v'' = 0$  is at 340 nm whereas the Franck-Condon Factors near 340 nm are vanishing small for  $v'' = 1$  or  $v'' = 2$ . In general, classical transition regions are determined by the Mulliken difference potential analysis [34]. The application of this classical analysis to femtosecond pump-probe experiments in molecular physics was discussed by our group in [20]. Because of the known average vibrational spacings ranging from 34, 4  $\text{cm}^{-1}$  at 337 nm ( $v' = 53 - 54$ ) to 28,4  $\text{cm}^{-1}$  at 343 nm ( $v' = 37 - 38$ ) we estimate a vibrational oscillation period of about 1 ps. The probe laser wavelength (540 nm) is chosen such, that only at the outer turning point a transition onto the repulsive  $2\Sigma_u^+$  state of  $\text{Na}_2^+$  is possible by energy and

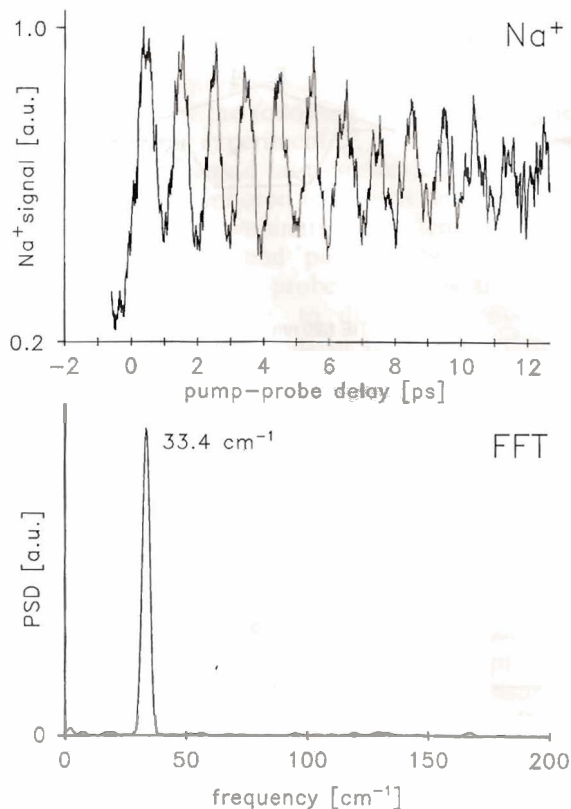
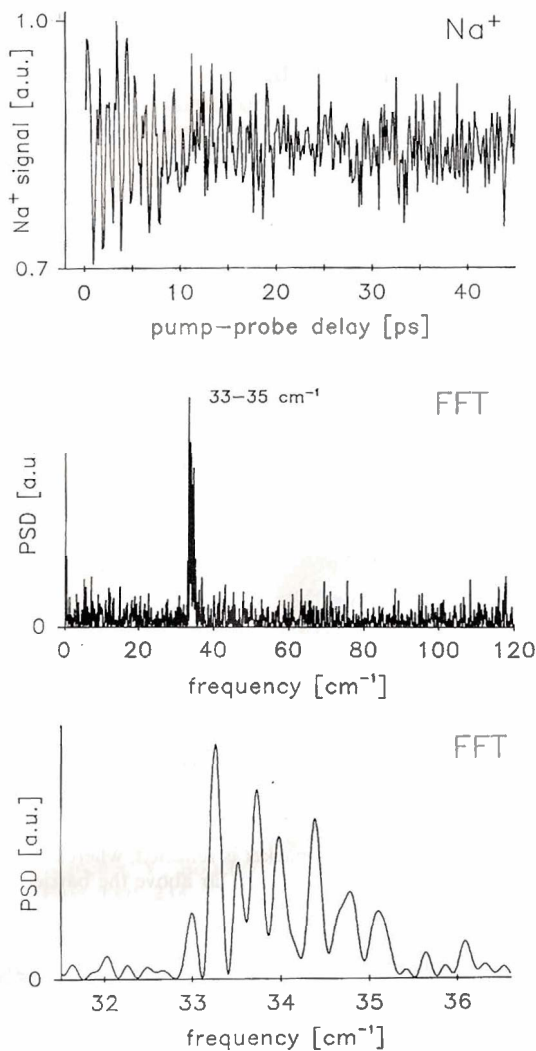


Fig. 3. The measured (upper part)  $\text{Na}^+$  fragmentation signal as a function of pump-probe delay time shows the oscillatory behaviour, determined by the  $(2) \ ^1\Sigma_u^+$ -potential curve. The lower part displays the Fourier transform of the transient  $\text{Na}^+$  spectrum. The individual energy spacings of the vibrational levels the wave packet is composed of are not resolved

Franck-Condon arguments. The probe laser will therefore form slow atomic  $\text{Na}^+$  fragments. By preparing the wave packet at the inner turning point of the  $(2) \ ^1\Sigma_u^+$  state and transferring it into the ionisation continuum at the outer turning point, the first signal maximum is observed only after half a vibrational period according to the formation of the ionic ( $\text{Na}^+$ ) fragments. Depending on the pump-probe delay the ionic ( $\text{Na}^+$ ) signal is expected to oscillate with a period of about 1 ps. This indeed is what we observe in the experiment. Figure 3 shows in the upper part the measured ionic ( $\text{Na}^+$ ) signal versus the pump-probe delay time. The 1 ps oscillation period is clearly seen. Due to the anharmonicity of the  $(2) \ ^1\Sigma_u^+$  potential the vibrational spacings are not constant within the spectral width of the exciting laser pulse. Therefore the amplitude of the observed oscillation decreases with time. This spreading of a vibrational wave packet takes place in a well defined way and the wave packet restores completely after a certain time, which is known as the recurrence time of a wave packet. The spreading and recurrence of a vibrational wave packet motion in the  $A \ ^1\Sigma_u^+$  state of the sodium dimer was earlier investigated experimentally in our group and compared with quantum simulations [23].

A Fast Fourier Transformation (FFT) analysis (lower part of Fig. 3) of the transient  $\text{Na}^+$  data displayed in the



**Fig. 4.** *Upper part:* First 40 ps of a 170 ps long  $\text{Na}^+$  transient obtained in the pump-probe scheme of Fig. 2. The spreading of the wave packet is clearly seen. *Middle part:* Fourier transformation of the transient  $\text{Na}^+$  signal. Only frequencies in the range of  $33\text{--}35\text{ cm}^{-1}$  contribute to the formation of the wave packet. *Lower part:* Enlargement of the Fourier transformation displayed in the upper part. The frequency distribution is composed of individual frequencies which correspond to the energy spacings of vibrational levels of the  $(2)^1\Sigma_u^+$  double minimum state forming the wave packet

upper part of Fig. 3 results in a frequency distribution centered around  $33.4\text{ cm}^{-1}$  (corresponding to 1ps), as expected from the spectroscopic data given above. The individual energy spacings of the vibrational levels the wave packet is composed of are not resolved. In order to obtain higher resolution in the frequency domain we increased the scan length in the time domain before applying the FFT analysis. The upper part of Fig. 4 shows the first 40 ps of a 170 ps  $\text{Na}^+$  transient obtained in the pump-probe scheme of Fig. 2. The decrease in amplitude of the oscillation with respect to pump-probe delay time is due to the spreading of the wave packet. The FFT analysis (middle part of Fig. 4) shows frequencies only in the range of  $33\text{--}35\text{ cm}^{-1}$ . This indicates that exclusively the  $(2)^1\Sigma_u^+$  state of the  $\text{Na}_2$  molecule is probed in this experiment. The vibrational frequencies of the  $\text{C } ^1\Pi_u$  state

**Table 1.** Vibrational energy spacings  $\Delta G(v)$  of the  $(2)^1\Sigma_u^+$  state of  $\text{Na}_2$  from femtosecond time domain data

$v'$	REMPI <sup>a</sup> [ $\text{cm}^{-1}$ ]	REMPI <sup>b</sup> [ $\text{cm}^{-1}$ ]	FTS <sup>c</sup> [ $\text{cm}^{-1}$ ]	fs-experiment [ $\text{cm}^{-1}$ ]
59–58	–	–	35.19	} 35.1
58–57	–	–	35.06	
57–56	35.1	35.5	34.91	
56–55	34.1	34.0	34.75	34.78
55–54	34.5	35.4	34.58	34.65 (s)
54–53	34.8	34.4	34.39	34.39
53–52	33.8	33.5	34.20	34.16 (s)
52–51	34.5	34.7	33.99	33.98
51–50	35.9	35.4	33.76	33.73
50–49	31.5	33.4	33.53	33.51
49–48	33.3	31.6	33.28	33.27
48–47	33.4	33.7	33.00	33.00
47–46	32.5	33.0	32.71	–
46–45	32.5	33.1	32.40	–
–	–	–	–	–
–	–	–	–	–
–	–	–	–	–

Vibrational energy spacings  $\Delta G(v)$  obtained from the FFT spectrum displayed in Fig. 4. The mark s denotes values obtained from shoulders in Fig. 4. The assignment of  $v'$  levels in the  $(2)^1\Sigma_u^+$  state is based upon high resolution Fourier transform spectroscopy (FTS) data. Results of nanosecond laser REMPI experiments are also given

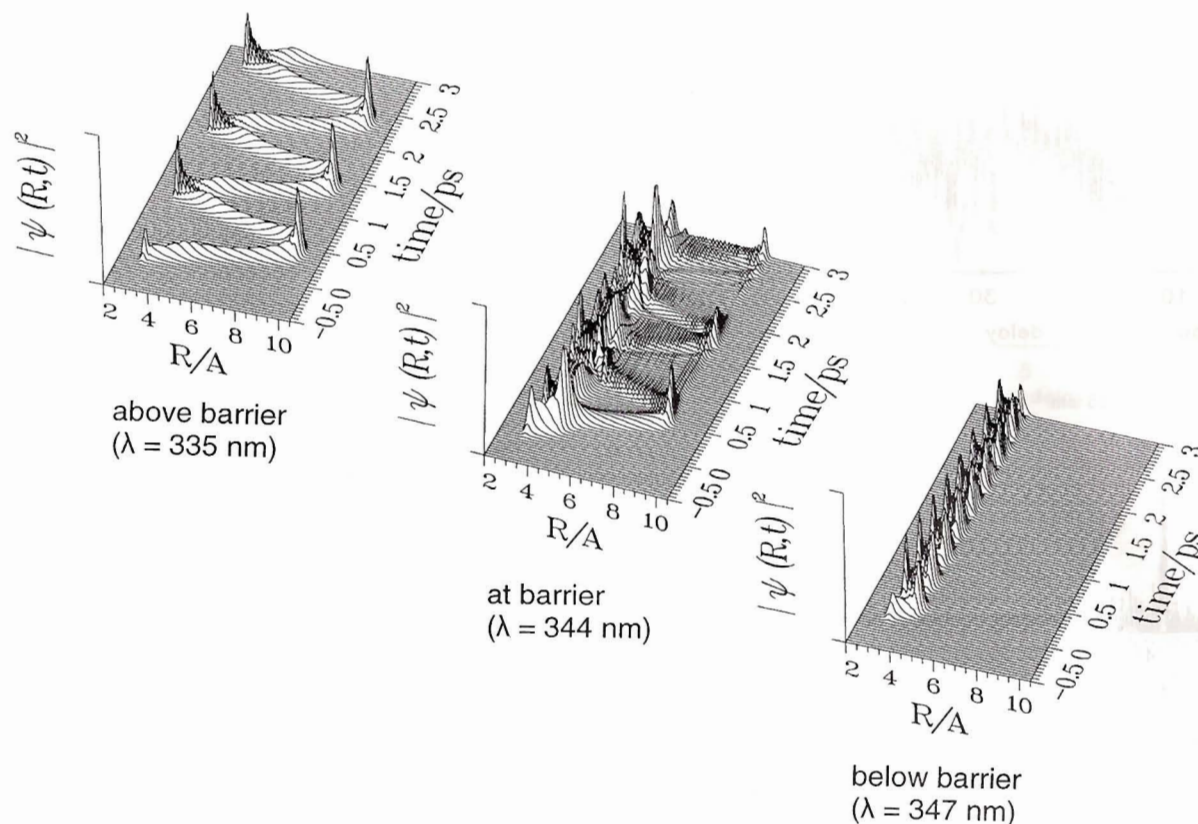
<sup>a</sup> Delacrétaz and Wöste [32]

<sup>b</sup> Haugstätter et al. [33]

<sup>c</sup> Cooper et al. [30]

( $T_e = 29621.37 \pm 0.5\text{ cm}^{-1}$ ,  $\omega_e = 116.25 \pm 0.2\text{ cm}^{-1}$  [30]) are not seen, because at 540 nm probe laser excitation no formation of slow  $\text{Na}^+$  fragments is possible out of the  $\text{C } ^1\Pi_u$  state for Frank-Condon reasons. Finally, the lower part of Fig. 4 shows this frequency range enlarged and individual vibrational frequencies are clearly seen. These frequencies are in fact the vibrational energy spacings the wave packet is composed of in the  $(2)^1\Sigma_u^+$ , as is confirmed by the comparison (Table 1) with high resolution frequency spectroscopy reported by Cooper et al. [30] and Vergès et al. [31]. The pump laser central wavelength belonging to this frequency range is 338 nm. The ns laser REMPI results of Delacrétaz and Wöste [32] and of Haugstätter et al. [33] are also shown for comparison. The accuracy of our data in comparison with the high resolution results of Cooper et al. is better than  $0.02\text{ cm}^{-1}$ .

As described in the introduction, the Tannor-Kosloff-Rice control scheme is based on controlling the duration of a wave packet on an excited electronic surface. This can be translated into classical terms as control of the internuclear distance. The feasibility of this idea is evident from the excitation scheme displayed in Fig. 2. As discussed above, the pump laser excites the molecule at the inner turning point, whereas the probe laser transfers the wave packet only at the outer turning point onto the repulsive  $^2\Sigma_u^+$   $\text{Na}_2^+$  state by energy and Franck-Condon reasons. During the propagation of the wave packet to the outer turning point the probe laser mainly produces molecular ions. In this set up, the control of internuclear distance by means of femtosecond pump-probe techniques leads either to ionisation or to ionisation induced fragmentation. The variation of the ratio of  $\text{Na}^+$  fragment ions



**Fig. 5.** Dynamical behaviour of a wave packet in the  $(2) \ ^1\Sigma_u^+$  double minimum state (Fig. 2) for different excitation energies (see text):  $\lambda_{\text{pump}} = 347.0 \text{ nm}$ : below the barrier;  $\lambda_{\text{pump}} = 344.2 \text{ nm}$ : at the

barrier - note that part of the wave packet is reflected, whereas the other part is transmitted;  $\lambda_{\text{pump}} = 335.0 \text{ nm}$ : far above the barrier

versus  $\text{Na}_2^+$  molecular ions, i.e. the control of ionisation induced fragmentation and ionisation as a function of internuclear distance will be discussed in detail in a forthcoming publication [22].

Future experimental work on this state will focus on the influence of the barrier on the vibrational wave packet motion. To illustrate the experiment we have simulated the propagation of a wave packet created at the inner turning point of the  $(2) \ ^1\Sigma_u^+$  state. Bandwidth limited laser pulses of 80 fs ( $\text{sech}^2$ ) were used in the simulations. The results are summarized in Fig. 5. A strong dependence of the wave packet dynamics with respect to its energy or equivalently to the wavelength of the exciting laser pulse is seen. At  $\lambda = 347 \text{ nm}$  the wave packet is excited below the barrier and therefore initially only motion within the inner well is possible. For longer evolution times we expect the wave packet to tunnel through the barrier. Centering the energy of the wave packet at the barrier ( $\lambda = 344.2 \text{ nm}$ ) the wave packet splits into a transmitted and a reflected part. At  $\lambda = 335 \text{ nm}$  the wave packet is far above the barrier and the dynamics represents nicely the classical motion of a point mass in this potential well. Transient electron spectra reflecting wave packet motion in this  $(2) \ ^1\Sigma_u^+$  state were calculated recently by Meier and Engel [35]. They used an excitation scheme similar to that described here. They showed that the spatial distribution of the probability density is directly mapped onto the electron spectra. The authors also pointed out, that the

coordinate dependence of the transition dipole moment between the  $(2) \ ^1\Sigma_u^+$   $\text{Na}_2$  and the  $^2\Sigma_g^+$   $\text{Na}_2^+$  state can be deduced from femtosecond pump-probe experiments.

#### 4 Conclusions

In this contribution we report on femtosecond time resolved pump-probe experiments on the  $(2) \ ^1\Sigma_u^+$  double minimum state of  $\text{Na}_2$ . The experiments were performed in a molecular beam with ion Time of Flight (TOF) detection. Several aspects of vibrational wave packet motion were investigated.

First we showed we can perform frequency spectroscopy using femtosecond pump-probe techniques in spite of the inherent spectral width of an ultrashort laser pulse. By Fast Fourier Transformation analysis of time domain data the vibrational energy spacings of the  $(2) \ ^1\Sigma_u^+$  double minimum well potential of  $\text{Na}_2$  were obtained with an accuracy of  $0.02 \text{ cm}^{-1}$  in comparison with high resolution Fourier transform spectroscopy results of Cooper et al. [30].

Secondly we demonstrated that the classical aspect of wave packet motion can be used in order to control the internuclear distance of the molecule. We pointed out that in a two color femtosecond pump-probe experiment the yield of the photo products  $\text{Na}_2^+$  versus  $\text{Na}^+$  as a function of pump-probe delay can be controlled. This type of

experiment is now referred to as "pump and control" experiment.

Finally we presented simulations of the influence of the potential barrier of the  $(2) ^1\Sigma_u^+$  state on the vibrational wave packet motion.

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