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13. ABSTRACT (Maximum 200 words)

A laser system based on a solid state injection seeded laser pumping optical parametric oscillators has been constructed for applications to molecule-surface interactions. This laser system generates pulsed, high-energy (10-100 mJ), broadly tunable, near-transform-limited linewidth radiation to be used for exciting molecular species to high vibrational levels. The approach selected mates an injection seeded Nd:YAG laser with spatially flat-topped pulses with an optical parametric oscillator operating in grazing incidence to generate the narrow bandwidth pulses. These pulses are then further amplified. A unique feature of the device is its frequency stabilization which is crucial for experiments in which molecules are maintained at high level of excitation for extended periods. A second laser system, based on a tunable, grazing incidence, dye laser is used to monitor excited molecules and products by laser ionization. Excellent performance has been obtained with this device, as demonstrated in experiments in which HCl in v=2, j=1 is scattered from surfaces.

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AFOSR Grant No. F49620-95-1-0089

Final Report  
December 1994 - December 1996

**HIGH-INTENSITY HIGH-RESOLUTION SOLID-STATE TUNABLE  
LASERS FOR INTERFACE CHEMISTRY**

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## I. EQUIPMENT, SETUP AND PROCEDURES

Funding was obtained to purchase and construct solid state *injection seeded* lasers based on optical parametric oscillators (OPO's) and Ti:S lasers that will generate pulsed, high energy (10-100 mJ), broadly tunable (200-2300 nm), *near-transform-limited-linewidth* radiation for use in studies of molecule-surface interactions that are relevant to materials interactions. The suggested approach was to design Nd:YAG pumped OPO's that will deliver near-transform limited linewidth, but will feature also frequency stabilization to enable long-term pumping of molecular species without drift. Each seed wavelength results in narrow linewidth signal *and* idler OPO outputs, which can also be frequency doubled and mixed. The laser system is used to impart high vibrational excitation to incident molecules and adsorbates. In particular the interactions of molecules with chemically significant vibrational energy with surfaces are examined.

In order to maximize versatility, a modular design was adopted that is advantageous for different kinds of experiments. In this approach, high-power Nd:YAG lasers with spatially flat-topped pulses pump a variety of devices that produce high-intensity IR beams. In applications where high-resolution or high-brightness is important, the plan was to use either single-mode high-resolution tunable diode lasers, or a grazing-incidence KTP OPO developed by Dr. Dean Guyer and now used by Continuum.

The pump lasers (Nd:YAG) were purchased from Continuum. One system was dedicated to OPO pumping, and the other was used to pump a dye laser providing the probe beam required to detect the highly excited molecules. Dr. Dean Guyer agreed to design and construct the grazing-incidence KTP OPO that serves as the oscillator stage in some of the applications. An information-exchange agreement with Continuum, the only manufacturer of high-resolution OPO's, has been reached regarding aspects of locking the lasers to single absorption lines. This is important in applications where the pump laser is parked on a wavelength coinciding with a narrow molecular absorption line. Dr. Matthew Johnson, a laser physicist who developed injection seeded OPO as a graduate student, has joined our team and assumed the primary responsibility for development.

The unique feature of this effort is the development of high-intensity narrow-bandwidth OPO with frequency stabilization which is described in some detail below.

The narrow-linewidth OPO is capable of high intensities in the IR and active frequency stabilization. The latter step is crucial to the success of our experiments which demand holding the pump frequency on a specific rovibrational level of the incident molecule for long periods of time. The OPO is based on an actively stabilized SLM KTP OPO tunable in the visible and near IR. This low-energy oscillator ( $\sim 0.5$  mJ signal + idler output) is amplified in a KTP OPA producing approximately 20 mJ total output. The frequency locking device is based on a novel design which includes monitoring the transmission of a temperature-stabilized etalon; it was developed for us by Dean Guyer (Laser Vision) with a license from Continuum.

Although quite complex, we are very pleased with the performance of the device. It has delivered consistently  $\sim 5$  mJ at  $1.7 \mu\text{m}$  with a bandwidth of  $0.01 \text{ cm}^{-1}$ . The frequency locking is stable for the several hours duration of the experiment. Since we are able to achieve saturation of the  $2 \leftarrow 0$  transition in HCl, we are very optimistic that higher overtones of HCl and overtones and combination bands of other molecules can also be pumped efficiently. It should be emphasized that the performance of narrow-bandwidth OPOs is crucially dependent on the beam shape and stability of the pump Nd:YAG laser, and our ability to purchase the best pump laser for this application, made possible by an AFOSR DURIP grant, was a prerequisite to our success.

The laser system was tested by studying the interaction of HCl with high internal energy on MgO(100). To this end beams of HCl in  $v=2, J=1$  were prepared by excitation at  $1.7 \mu\text{m}$ . With the newly developed OPO, saturation of  $v=2$  was achieved, and moreover the temporal profile of the beam was reduced to  $\sim 200$  ns, which enables us to carry much better time-of-flight distributions unraveling energy transfer mechanisms. The FWHM of  $\sim 200$  ns is limited by the spot size of the pump laser. The good temporal resolution in the incident beam allows us to better resolve the time profiles of the state-selected scattered beam. By controlling the incident energy we can see changes in the TOF curves corresponding to a transition from direct inelastic

scattering to a mechanism dominated by transient trapping and desorption. These results are progressing at a fast pace, with the S/N ratio improving sharply as we gain more experience with the new OPO and following the installation of the high-intensity probe laser.

## II. PERSONNEL

Mikhail Korolik — graduate student

Don Arnold — postdoctoral fellow

Matthew Johnson - postdoctoral fellow, supported by NSF

Minda Suchan - graduate student, supported partly by a Dean Fellowship

Dean Guyer — consultant

## III. INTERACTIONS/TRANSITIONS

### A. PRESENTATIONS AT MEETINGS

1. "Molecule-surface collision-induced dissociation of highly excited NO<sub>2</sub> on MgO(100)", J. Singleton, D.W. Arnold, M. Korolik, H. Reisler and C. Wittig, ACS Meeting, Anaheim, April, 1995.
2. "Molecule-surface collision-induced dissociation of highly excited NO<sub>2</sub> on MgO(100)," D.W. Arnold, M. Korolik, H. Reisler and C. Wittig, 1995 Conference on the Dynamics of Molecular Collisions, Asilomar, CA, July 1995.
3. "Reactive and inelastic processes in molecule-surface interactions," D.W. Arnold, M. Korolik, M. Johnson, H. Reisler and C. Wittig, invited talk, ACS Meeting, Orlando, FL, August 1996.
4. "Inelastic scattering of HCl from MgO(100)," D.W. Arnold, M. Korolik, M. Johnson, H. Reisler and C. Wittig, Poster presentation, ACS Meeting, Orlando, FL, August 1996.
5. "Inelastic scattering of HCl( $v=2, J+1$ ) from MgO(100)," D.W. Arnold, M. Korolik, M. Johnson, M. Suchan, H. Reisler and C. Wittig, Poster presentation, Gordon Conference on Molecular Energy Transfer, Ventura, CA, Jan. 1997.

## B. INTERACTIONS WITH OTHER LABS

1. Following a recent visit to our labs by representatives from Continuum, Inc., it is our understanding that they will further develop and market the OPO presently used in the experiments reported herein. If so, this will constitute a significant contribution to the realm technology transfer. Namely, we were the initial test site for the development stage of this device.
2. Curt Wittig visited the Phillips Laboratories (Dr. Ed Murad) and gave a seminar.
3. Curt Wittig visited Aerodyne (C. Kolb, D. Worsnop) to discuss interactions of halogen containing compounds on sapphire and alumina surfaces and gave a talk.
4. H. Reisler and C. Wittig participated in a workshop on interactions of alumina with the atmosphere, organized by John Lamb TRW, May 1996.

## VI. HONORS/AWARDS

Hanna Reisler received the 1994 Max Planck Research Award conferred jointly by the Max Planck and Alexander von Humboldt Societies.

Hanna Reisler was elected Fellow of the American Physical Society.