論文の内容の要旨

論文題目 : Laser-field-free and field-free orientation of state-selected molecules
（状態選別した分子のレーザー電場のない条件下、及び完全に外場のない条件下での配向制御）

氏名 : 文 堤會

Nowadays, thanks to the advance of laser technologies, molecular alignment has matured to a state with a number of associated studies. One of the most widely adopted techniques in various experiments associated with gas phase molecules is "impulsive alignment" technique. When molecules are irradiated with a linearly polarized impulsive laser field, the angular distribution of the molecules are synchronized along the laser field. In fact, the impulsive alignment technique has broadened the scope of spatially aligned molecules from the original focus on chemical reaction dynamics to new applications such as optimal control of multiphoton ionization and molecular imaging based on high harmonics generation.

The control and manipulation of the directional features of molecules, even though a long history of it, is still very promising research area. Especially, development of a robust orientation technique is an important subject of research and many scientists around the world have been developing it. Furthermore, considering that a lot of studies of field-free molecular alignment and orientation are restricted on the molecules with simpler structures and samples of oriented asymmetric top molecules are mainly created in the presence of laser field, researches on angular confinement of molecules will have been an attracting subject until a technique for controlling molecular orientation of large asymmetric top molecules in the field-free condition is established.

We have achieved laser-field-free, strong molecular orientation of asymmetric top molecules in the presence of a weak electrostatic field for the first time. One necessary condition for controlling rotational motion of a gas phase molecule is creating a rotational superposition of free-rotational states. The generated rotational wave packet must be in a good coherent condition for the angular confinement of the molecule. On the other hand, because of the complicated rotational spectrum of the asymmetric top molecule, making a good coherent wave-packet for field-free, strong alignment and orientation is a much more difficult subject compared to that of molecules with simpler structures like linear and symmetric top molecules.

For strong, laser-field-free orientation of asymmetric top molecules, we have employed plasma shutter technique for shaping a laser pulse. With the plasma shutter, a special laser pulse with a slow turn on and a rapid turn off is shaped. The shaped pulse works as a tool for achieving strong molecular orientation of molecules in the laser-field-free condition. The achievable degrees of orientation for initial single rotational state are maximized, when the external fields are applied slowly so that the interaction time scale is close to the adiabatic limit, while the achievable degrees of alignment do not critically depend on the interaction time scale. The oriented rotational states created by the slow turn on can be transferred into laser-field-free states by the rapid turn off.

Moreover, for further improvement of the degrees of orientation, we employed a molecular sample which is rotational-state-selected by Stark acceleration. When molecules pass through a spatially inhomogeneous strong electrostatic field, they experience a force depending on Stark energy shifts of individual rotational states. Accordingly, the inhomogeneous electrostatic field can be used for the state-selection of molecules. Since some of rotational states irradiated with external field(s) are oriented one way and the others are oriented the other way, the degree of orientation that we get from a group of molecules is much lower than that from a certain rotational state. Therefore, we have employed molecules rotational-state-selected by a molecular deflector and achieved much higher degrees of orientation than that achieved from a thermal ensemble of molecules. With this combination of the two techniques of the plasma shutter and the rotational-state selection, a state-of-the-art orientation technique has been established.

On the other hand, looking at obtained experimental results from the asymmetric top molecules, we have observed persistent alignment and orientation for 5-10 ps after the rapid turn off of the laser field by plasma shutter. Such a remarkably slow dephasing of coherent wave-packet has never been observed, which motivated us to the underlying physics by numerical simulations. We have numerically found that, even though a laser pulse whose pulse width is much longer than the rotational period of the molecule, a nonadiabatic effect plays an important role in determining the rotational dynamics of molecules in usually conceivable experimental conditions.

For the explanation of the numerical method that we have developed, that is quite similar to the time-dependent unitary perturbation method, a large part of the present thesis is dedicated. With this numerical method, most of the simulations pertaining to the rotational dynamics of molecules have become possible. Especially, we can describe the nonadiabatic effect caused in the regime where the interaction between large asymmetric top molecules and external field is much longer than rotational period of the molecule but not long enough to ensure completely adiabatic process.

Our development of the numerical method is pertaining to the advance of the orientation technique, which is evidenced by a fact that experimental achievements for angular controlling of

molecules have been accompanied by theoretical efforts to understand and estimate rotational dynamics,

After the laser–field–free orientation technique in the presence of a weak electro–static field has been developed, we have tried to accomplish completely field–free orientation to make an oriented molecular sample available even without an electrostatic field. We have employed a two–color laser pulse with a slow turn on and a rapid turn off, for the field–free molecular orientation. However, we have found experimentally and theoretically that the orientation process on the slow turn on of the laser pulse is not close to adiabatic, because of the difference in the pulse width between the fundamental and the second–harmonic laser pulses. Thus, degree of orientation expected to be achieved by adiabatic interaction can not be achieved this time. In spite of such a difficulty associated with the orientation technique, the state–selection technique and the plasma shutter technique have attributed to the field–free strong orientation of OCS molecules.