

Contents

List of Figures	19
List of Tables	25
I Preface	27
1 Introduction	29
1.1 Aim and motivation	29
1.2 Molecular interactions with fields: Origin	30
1.2.1 Molecular alignment and orientation	30
1.2.2 Controlling translational motion of diatomic molecules with external fields	33
1.3 Experimental applications of molecular interactions with fields . . .	35
1.3.1 Molecular traps	35
1.3.2 Cold and Ultracold Molecules	35
1.3.3 Chemical reaction dynamics	36
1.3.4 Quantum Computation with Polar molecules	37
1.3.5 Other applications	39
1.4 Scope and outline of the present thesis	40

II	Theory	45
2	Rotational structure of diatomic molecules	47
2.1	Rigid rotor	48
2.2	Symmetric Top	50
2.3	Angular momentum algebra	51
2.4	Spin angular momentum	53
2.5	Hund's coupling cases	55
2.5.1	Hund's case (a)	55
2.5.2	Hund's case (b)	57
2.5.3	Hund's case (c)	57
2.5.4	Hund's case (d)	57
2.5.5	Hund's case (e)	58
2.6	$^2\Sigma$ molecule	59
3	Interaction of $^2\Sigma$ with external fields	61
3.1	Interaction with an electric field	61
3.2	Interaction with a magnetic field	63
3.3	Interaction with an optical field	64
3.4	Interaction of nuclear spin	66
4	Two molecule interactions	69
4.1	Electric dipole-dipole interactions	69
4.1.1	Approximate solution of electric dipole-dipole interaction	71
4.1.2	Exact form of electric dipole-dipole interaction and it's matrix elements	71
III	A single molecule in fields	73
5	Single-Field Effects	79
5.1	Pure Stark interaction	79

5.2	Pure Zeeman interaction	82
5.3	Pure polarizability interaction with an optical field	85
6	Double-Field Effects	89
6.1	Congruent electric and optical fields	89
6.1.1	As a function of optical field	89
6.1.2	As a function of electric field	94
6.2	Congruent electric and magnetic fields	95
6.2.1	As a function of magnetic field	95
6.2.2	As a function of electric field	96
6.3	Congruent magnetic and optical fields	104
6.3.1	As a function of magnetic field	104
6.3.2	As a function of optical field	104
7	Triple-Field Effects	107
7.1	Variation of the electric field	107
7.2	Variation of the magnetic field	114
7.3	Variation of the optical field	115
IV	A pair of molecules in fields	119
8	Pair eigenstates in magnetic field	125
8.1	Pair-eigenstates in the absence of the electric dipole-dipole coupling, $\Xi = 0$	125
8.2	Pair-eigenstates in the presence of a small dipole-dipole coupling, $\Xi \ll 1$	128
8.3	Pair-eigenstates in the presence of large dipole-dipole coupling, $\Xi \leq 1$	131
9	Mutual alignment of a pair of coupled rotors	135
9.1	Analytic model of pairwise alignment	136

V	Quantum Computer	141
10	A unit quantum circuit: A pair of polar $^2\Sigma$ molecules	147
10.1	Choice of qubits	147
10.2	Behavior of a two-qubit system in concurrent electric and magnetic fields	149
11	CNOT gate implementation schemes	153
11.1	Scheme I	154
11.2	Scheme II	154
12	Simulations of CNOT gate for a pair of NaO molecule	159
12.1	Scheme II for a pair of NaO molecules	160
12.2	Feasibility of Schemes I and II	164
12.2.1	Broadening	164
12.2.2	Multiple gate operations	167
VI	Conclusions	169
13	Summary and outlook	171
13.1	Summary	171
13.2	Outlook	175
VII	Appendix	177
A	Adiabatic following of states using the dot product	179
B	Normalization of the wavefunctions and its consequences for the plotted probability distributions	181
C	Direction cosine matrix elements in the symmetric top basis set	185
D	Matrix elements in Hund's case (a) basis set	189

E	Matrix elements of the pairwise alignment cosine in the cross product basis set of the two molecules	193
F	Matrix elements of the electric dipole-dipole operator in the cross product basis set of the two molecules	195
G	Conversion factors	199
	Bibliography	201