Desorption due to recoil induced by neutrino emission and auger relaxation of 37Cl following the electron capture decay of 37Ar

by Lin Zhu

A thesis submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Physics

Montana State University

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Abstract:

A novel experiment was developed in this work to study the desorption and the Auger relaxation processes of 37Cl following the 37Ar electron capture decay. For the first time, the desorption of 37Cl ions due to recoil induced by neutrino emission in this decay process was observed. The kinetic energy distribution of the desorbing 37Cl ions was accurately measured by using coincidence techniques. The resulting 37Cl ion energy ranges from 5 eV to 13 eV with a maximum at around 9 eV and an FWHM about 3 eV. The charge state distribution of the desorbing 37Cl ions was also measured. The resulting charge state distribution is: 53% of the total ions have charge +e, 21% have charge +2e and 26% have charge +ne, where $n \geq 3$. The desorbing probability of 37Cl ions was measured by two independent experiments which gave the result of 9.4±1.2%. The energy distribution, the charge state distribution and the desorbing probability of 37Cl ions are all quite different as compared with the expected values for an isolated Cl atom. These differences are explained in the desorption model involving charge exchange and Coulomb repulsion between 37Cl ions and their surrounding atoms.

The electron capture decay also creates a highly unusual initial state in the 37Cl atom which allows direct observation of some novel relaxation processes which are. amenable to many body theory, but essentially impossible to probe experimentally with conventional techniques. For the first time, direct evidence of the double Auget decay of a K-hole and the large shift in energy (22 eV) of an LMM Auger line was reported. The double Auger decay probability and energy distribution of the two double Auger electrons were measured by using coincidence techniques. The resulting double Auger decay probability ranges from 12±0.3% to 15±0.4% of the total Auger decay, The preferred energy distribution of the double Auger emission is for one of the electrons to take most of the energy, with the second receiving the small remaining balance. A model explaining the large shift in energy of an LMM Auger line is also provided.

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of

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in

Physics

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APPROVAL

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This thesis has been read by each member of the thesis committee and has been found to be satisfactory regarding content, English usage, format, citations, bibliographic style, and consistency, and is ready for submission to the College of Graduate Studies.

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DEDICATED TO MY GRANDMOTHER
AND MY PARENTS

VITA

Lin Zhu was born on October 29, 1962 in Beijing, China to Shijun Zhu and Wenjuan Xiang. He has a younger sister, Hong Zhu.

Lin Zhu was raised in the beautiful campus of Tsinghua University, Beijing China where he attended the university-attached primary school in 1969, junior high school in 1974 and high school in 1977. In 1980, he was admitted to Tsinghua University, one of the best schools in China. He completed his undergraduate education with a B.S. degree in electrical engineering in 1985. In the same year, he started his graduate study in the Graduate College of Tsinghua University. From 1987 to 1989, he studied in the Physics Department of the Technical University of Munich, Munich, West Germany. In 1989, he came to the wonderful small town called Bozeman. He got his M.S. degree in physics from Montana State University in 1992.

PREFACE

Eight years ago, when Dr. Avci, a surface scientist met Dr. Hindi, a nuclear physicist half a world away in Saudi Arabia, they came up with a brilliant idea: using surface analysis techniques to study nuclear reactions. Six years ago, Dr. Menzel invited me to his office, "I just talked to Prof. Lapeyre in Montana. You should not disappoint them when you get there," he told me in German, "because I have just put up a lot of good words for you." Four years ago, there began this research project. With the help of a German professor, I got the opportunity to work under the guidance of professors with Turkish, Lebanese and American origin. I would like to dedicate this report to these mentors who have shown me how small this world could be for physicists.

My sincere thanks to those who made this possible. To Dr. Lapeyre for his help in my coming to this great country, for his continuous support, supervision and confidence in me. To Dr. Avci for his scientific inspiration, encouragement and patient guidance throughout this thesis work. To Dr. Hindi who has helped me throughout this project with his sharp instincts in physics and tireless working spirits. To Dr. Anderson for his important criticisms and advice that have been and will remain as a beneficial experience for me. To Dr. Smith, Dr. Tuthill and Department Chairman, Dr. Hermanson for their great support in my graduate study. To the wonderful staff, Alice, Margaret and Rose for their cheerful help. To Erik and Norm for their excellent technical support.

TABLE OF CONTENTS

Page

VITA v

PREFACE vi

TABLE OF CONTENTS vii

LIST OF TABLES x

LIST OF FIGURES xi

ABSTRACT xv

1. INTRODUCTION 1

Concepts of the Work 2

Desorption due to Recoil Induced by Neutrino Emission 2

Surface Interactions of ^{37}Cl Ion 5

Auger Relaxation of ^{37}Cl Atom 8

Historical Review of Related Works 10

2. THEORY 14

Recoil Induced by Neutrino Emission 14

Models on Stimulated Desorption 17

The Menzel-Gomer-Redhead (MGR) Model 18

TABLE OF CONTENTS—Continued

Page

The Knotek-Feibelman (KF) Model 22

Desorption Due to "Coulomb Explosion" 25

Theory of Auger Transition 29

Auger Effect 29

Auger Energies and Intensities 33

Double Auger Transition 38

3. EXPERIMENT 45

Experimental Setups 45

The UHV System 46

The Substrates 49

The Detectors 51

The Position Sensitive Detector 56

The Radioactive ^{37}Ar Sources 61

Coincidence Techniques and Electronic Systems 66

Experimental Procedure 73

4. RESULTS AND DISCUSSIONS 77

Desorption due to Recoil Induced by Neutrino Emission 77

Energy Distribution of ^{37}Cl Ions 77

Charge-State Distribution of ^{37}Cl Ions 83

TABLE OF CONTENTS—Continued

Page

Results on Multiple and Mixed Layers.....91

Desorption Probability of ^{37}Cl Ions..... 95

Discussion of the Desorption Results..... 99

Auger Relaxation of a ^{37}Cl atom.....108

^{37}Cl LLM and LMM Auger Peaks..... 109

Discussion of ^{37}Cl LMM Auger Results.....113

Double Auger Decay of a ^{37}Cl atom..... 117

^{37}Cl Double Auger Probability..... 118

^{37}Cl Double Auger Energy Distribution.....127

Discussion of ^{37}Cl Double Auger Decay Results..... 133

5. CONCLUSIONS.....140

Conclusions.....140

Perspective on Neutrino Mass Limit Study..... 143

REFERENCES..... 146

APPENDICES..... 156

Appendix A: Introduction to Neutrino Physics and Neutrino Mass Limit Study..... 156

Appendix B: Designing Diagrams of the Detectors..... 168

Appendix C: Plasmon Decay into Multi-Electron-Hole Pairs in Si(111)... 176

LIST OF TABLES

Table		Page
2.1	Table of the Energies, Symbols and Number of Electrons for Ar and Cl Core Levels	29
2.2	Table of Final States of Ar KLL Auger Series	32
2.3	Table of Ar LMM Auger Energies and Relative Intensities	36
2.4(a)	Table of Auger Transition Probabilities for a K-hole Vacancy	36
2.4(b)	Table of Auger Transition Probabilities for L-hole Vacancies	37
2.5	Table of Final Charge State Distribution of Ar after Auger Transition of an 1s Initial Hole	38
4.1	Table of Charge State Distribution Results from Experiments and Simulations	84
A1.1	Table of the Three Generations of Quarks and Leptons	161
A1.2	Table of Neutrino Mass Fitting Results	165

LIST OF FIGURES

Figure		Page
1.1	Electron capture decay and ^{37}Cl ions desorption in $^{37}Ar \rightarrow ^{37}Cl + \nu$	3
1.2	The original 9.5 eV recoil energy and the additional energy due to charge exchange and Coulomb repulsion of the ^{37}Cl ions	6
1.3	The unusual initial state of ^{37}Cl atom from EC decay	9
1.4	Normal Auger transition and double Auger transition	10
2.1	The energy level scheme of MGR model	19
2.2	The energy level scheme of Antoniewicz model	21
2.3	The KF model for O^+ ion desorption	23
2.4	Schematic displaying terms of a Hubbard Hamiltonian with intrasite Coulomb term U_{eff}	27
2.5	KLL Auger transition and LLM Coster-Koing Auger transition	30
2.6	Energy changes of an Auger electron entering analyzer with work function	34
2.7	Relative intensities of Auger peaks of Ar with 1s hole calculated by using RACS	37
2.8	Brueckner-Goldstone diagrams for the partial amplitudes of double Auger decay	40
2.9	Diagrams for double Auger decay models	42
2.10	Double Auger energy distribution calculated for atomic Ne	43
3.1	UHV system schematics	47

LIST OF FIGURES—Continued

Figure		Page
3.2	Auger peaks of Ar and Carbon from the substrates at different temperatures	48
3.3	TPD spectrum of Ar from the graphite substrate	50
3.4	The mounting bridge between the graphite substrate and the cold finger	51
3.5	Diagram of the home-made channeltron based detector	53
3.6	Diagram of the home-made microchannel plates based detector	55
3.7	MCP pulse high distribution and detecting efficiencies for electron and ion	55
3.8	Geometry of a circular arc terminated resistive anode encoder	59
3.9	Diagram of the home-made MCP-RAE based PSD detector	60
3.10	The gamma ray spectrum of the radioactive gas source taken by an HP-Ge detector	62
3.11	The gamma ray spectrum of the ^{37}Ar gas source	63
3.12	Double sealing ^{40}Ca in quartz ampoules before nuclear irradiation	65
3.13	Schematic diagram of the first coincidence measurement system	67
3.14	True coincidence counts and accidental coincidence counts	68
3.15	Schematic diagram of the second coincidence measurement system	70
3.16	The logic block diagram of the second coincidence measurement system	71
4.1	The Monte-Carlo simulation of the ^{37}Cl ion recoil velocity distribution	79

LIST OF FIGURES—Continued

Figure		Page
4.2	Time-of-flight spectrum obtained by using coincidence measurement and the corresponding energy spectrum of the desorbing ^{37}Cl ion	81
4.3	Comparison of ToF spectra between the Monte-Carlo simulations and the experimental results	82
4.4	Using retarding field energy analyzer to measure charge state distribution	85
4.5	Retarding field energy spectrum of the desorbing ^{37}Cl ion	86
4.6	Calculating charge state distribution from the RFE spectrum	87
4.7	Comparison of charge state distribution and the REF spectra between the Monte-Carlo simulation and the experimental results	89
4.8	Comparison of the maximum kinetic energy obtained by ToF and RFE spectra	90
4.9	RFE spectra of desorbing ^{37}Cl ion at different coverage	92
4.10	Comparison of RFE spectra, total ion counts and maximum kinetic energies among different mixtures of radioactive and stable Ar	93
4.11	Comparison of energy distribution of desorbing ^{37}Cl ion from different coverage	95
4.12	KLL and LMM Auger electron-electron coincidence ToF spectrum	98
4.13	Local environment of ^{37}Cl ion	100
4.14	The simulation results of Cl^+ ion exchange charge with metal surface	104
4.15	The model explaining charge exchange and Coulomb repulsion during ^{37}Cl ion desorption	106
4.16	The RAE spectrum of ^{37}Cl LMM Auger	110

LIST OF FIGURES—Continued

Figure		Page
4.17	Comparison of LMM Auger spectra among ^{37}Cl following EC decay, Cl from CsCl and physisorbed ^{40}Ar	111
4.18	^{37}Cl LMM Auger spectrum measured with new radioactive source	113
4.19	Comparison between ^{37}Cl LMM Auger spectrum and Cl conventional Auger lines	114
4.20	The Auger cascade model explaining the LMM^{H} peak	116
4.21	Spectrum of coincidence measurement between KLL Auger and LMM Auger	119
4.22	Spectrum of coincidence measurement between two Double Auger electrons	120
4.23	The RFE spectrum of ^{37}Cl LMM Auger by the MCP detector	123
4.24	Spectra of coincidence measurement for KLL and LMM Auger electrons with different retarding screen voltages	126
4.25	Spectra of coincidence measurement for double Auger electrons with different retarding screen voltages	128
4.26	Double Auger energy distribution	130
4.27	Differential energy dependence of the double Auger probability as compared with theoretical prediction	131
4.28	Double Auger energy distribution measured with the new source	132
4.29	Schematic diagram of a triple coincidence spectrum	134
4.30	Comparison of double Auger coincidence spectra between experimental and simulation results	138

ABSTRACT

A novel experiment was developed in this work to study the desorption and the Auger relaxation processes of ^{37}Cl following the ^{37}Ar electron capture decay. For the first time, the desorption of ^{37}Cl ions due to recoil induced by neutrino emission in this decay process was observed. The kinetic energy distribution of the desorbing ^{37}Cl ions was accurately measured by using coincidence techniques. The resulting ^{37}Cl ion energy ranges from 5 eV to 13 eV with a maximum at around 9 eV and an FWHM about 3 eV. The charge state distribution of the desorbing ^{37}Cl ions was also measured. The resulting charge state distribution is: 53% of the total ions have charge +e, 21% have charge +2e and 26% have charge +ne, where $n \geq 3$. The desorbing probability of ^{37}Cl ions was measured by two independent experiments which gave the result of 9.4±1.2%. The energy distribution, the charge state distribution and the desorbing probability of ^{37}Cl ions are all quite different as compared with the expected values for an isolated Cl atom. These differences are explained in the desorption model involving charge exchange and Coulomb repulsion between ^{37}Cl ions and their surrounding atoms.

The electron capture decay also creates a highly unusual initial state in the ^{37}Cl atom which allows direct observation of some novel relaxation processes which are amenable to many body theory, but essentially impossible to probe experimentally with conventional techniques. For the first time, *direct* evidence of the double Auger decay of a K-hole and the large shift in energy (22 eV) of an LMM Auger line was reported. The double Auger decay probability and energy distribution of the two double Auger electrons were measured by using coincidence techniques. The resulting double Auger decay probability ranges from 12±0.3% to 15±0.4% of the total Auger decay. The preferred energy distribution of the double Auger emission is for one of the electrons to take most of the energy, with the second receiving the small remaining balance. A model explaining the large shift in energy of an LMM Auger line is also provided.

CHAPTER 1

INTRODUCTION

On February 23, 1987, a supernova burst in the Large Magellanic Cloud (SN1987a) and sent rays of neutrinos to the earth. The sudden release of the 10^{53} erg of energy from the dying star triggered merely 100 or so "neutrino" counts in huge underground detectors around the world. This, however, caused enough excitement for several groups of physicists—the burst of SN1987a gave them a golden opportunity to study the mysteries of neutrinos.[1,2] Among these mysteries, the biggest one is whether neutrino has a rest mass or not, and what that mass might be. The neutrino mass problem is one of the most fundamental questions in physics. The answer to this question could have profound implications for particle physics, astrophysics and cosmology.[3]

The mass information of a neutrino can be obtained from a nuclear reaction involving the creation or annihilation of a neutrino.[1-3] One of these kinds of nuclear reactions is the electron capture (EC) decay. In this thesis, the desorption due to recoil induced by neutrino emission and the Auger relaxation processes of ^{37}Cl following the EC decay of ^{37}Ar is reported. It is demonstrated in this work that the EC decay of ^{37}Ar provides not only a possible approach to the neutrino mass problem but also a rare

opportunity for some novel studies in surface and atomic physics.[4-7] The focal point of this thesis is to report the results of these novel studies.

The problems addressed in this thesis work fall into two categories: The first one is the study of the desorption due to recoil induced by neutrino emission in the EC decay process, $^{37}Ar \rightarrow {}^{37}Cl + \nu$, where ν is a neutrino. In this EC decay process, could the emission of the neutrino cause enough recoil of the adsorbed daughter atom ^{37}Cl to break the absorption bond? If the desorption does occur, what factors would affect the energy distribution of those desorbing daughter atoms? The second category is the study of the Auger relaxation processes of the ^{37}Cl daughter atom. The EC decay creates a deep vacancy in the energy levels of the ^{37}Cl atom while the whole atom is still in a neutral charge state. As discussed later, this initial state is highly unusual as compared with those commonly encountered initial excitations. What are the relaxation processes or the decay channels associated with this particular initial state? To better understand these questions, the concepts of these studies will be introduced first in the following paragraphs.

Concepts of the Work

Desorption due to Recoil Induced by Neutrino Emission

The first part of the study is to measure the spectrum of recoil velocities of ^{37}Cl ions following the EC decay of ^{37}Ar physisorbed on a well defined surface. In this EC

decay process which converts ^{37}Ar into ^{37}Cl, a proton in the nuclei of ^{37}Ar captures a 1s or 2s electron and becomes a neutron, while a neutrino is emitted and the ^{37}Cl daughter atom recoils in the direction opposite to that in which the neutrino moves. This recoil may break the bond between the ^{37}Cl atom and the substrate and cause desorption of the ^{37}Cl ions from the surface (Fig. 1.1). The initial 1s or 2s vacancy created by the EC process will decay through Auger cascades or x-ray emission. Therefore the daughter ^{37}Cl atom is usually multiple ionized after the EC decay.

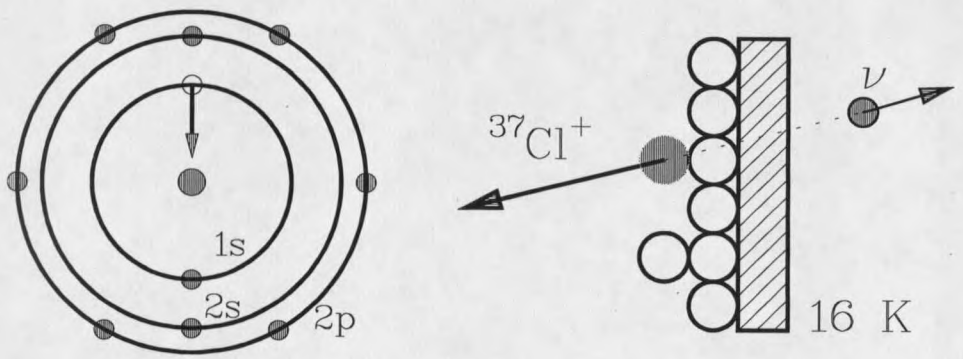

Figure 1.1. (a) Electron is captured from a low (1s) orbital: $^{37}\text{Ar} \rightarrow {}^{37}\text{Cl} + \nu$. (b) ^{37}Cl ion recoils and desorbs from surface due to neutrino emission.

For an isolated ^{37}Ar atom, the recoil process is a two-body problem with a total reaction energy, Q = 814 keV (Ref. 8). Energy and momentum conservation dictates that

the recoil energy must be 9.54 eV, if a massless neutrino with about 814 keV of energy is emitted. If a neutrino with mass m_ν is also emitted in a certain fraction of the EC decays, then the corresponding recoiling ions will have lower kinetic energy. The fractional change in recoil kinetic energy is $\Delta E/E = (m_\nu/Q)^2$ (Ref. 3), where Q, the total reaction energy is the rest mass (mc^2) difference between ^{37}Ar and ^{37}Cl. Thus the heaviest massive neutrino that could be emitted in this EC decay would have a rest mass of 814 keV. If a 250-keV neutrino were to be emitted, for example, the fractional change in recoil energy of the ^{37}Cl would be 9.4%. The lowest neutrino mass to which this method could be sensitive depends on the ability to resolve the peak associated with the emission of the massive neutrino from the dominant peak associated with the emission of the massless neutrino in the recoil spectrum.

There are well known and calculable effects that would contribute to the broadening in the recoil energy spectrum for an isolated ^{37}Ar atom. Some of these effects can be studied by computer simulations. However, for an ^{37}Ar atom physisorbed on a cold surface, and possibly surrounded by other Ar atoms, it is not obvious whether the recoil due to neutrino emission could break the bond between ^{37}Cl atom and the substrate and cause desorption. Even if the desorption does occur, it is still much more difficult, as compared to the isolated ^{37}Ar case, to determine what effects would contribute to the broadening in the recoil energy spectrum. To approach these questions, it is necessary to examine the surface interactions during the ^{37}Cl ion desorption process.

Surface Interactions of ^{37}Cl Ion

Adsorption and stimulated desorption have been very useful tools to study surfaces. Since the 1960s, several models have been developed to describe the interactions between adsorbates and the substrates. Current models involve detailed knowledge of surface binding energies, adsorption site symmetries and potentials, substrate phonon spectra, and surface electronic structures.[9-12] The neutrino recoil in this work provides an entirely new way to study the desorption dynamics. A simple model of the desorption in this case is described as follows: First, the adsorbed ^{37}Ar atoms undergo EC decay and become ^{37}Cl ions with 9.54 eV recoil energy to move away from the surface. Before these ^{37}Cl ions can leave the surface, the interactions between the ^{37}Cl ions and the substrate will result in additional kinetic energies on the top of the 9.54 eV recoil energy. These additional energies will contribute to the broadening of the recoil energy spectrum. One of the major goals in this work is to understand these interactions.

The first interaction to be discussed is the bonding between ^{37}Ar and the cold substrates. The ^{37}Ar atom is physisorbed on gold or graphite substrates. At the critical temperature of 65 K Ar begins to form a monolayer on these substrates. By about 30 K, multiple adsorbed layers (Ar ice) are formed.[13-15] It has been found that these multiple adsorbed layers are likely to form a face-centered cubic (fcc) polycrystalline layer at the experimental temperature of 16 K.[16] Therefore the adsorbed Ar could actually form a band structure and solid state physics has to be taken into account. The bonding between

an ^{37}Ar atom and the substrate or the neighboring Ar atoms is established through the van der Waals interaction which gives rise to a binding energy of about 100 meV.[15] This interaction cannot play a major role in the broadening of the recoil energy spectrum because the 100 meV interaction energy is much less than the 9.54 eV recoil energy.

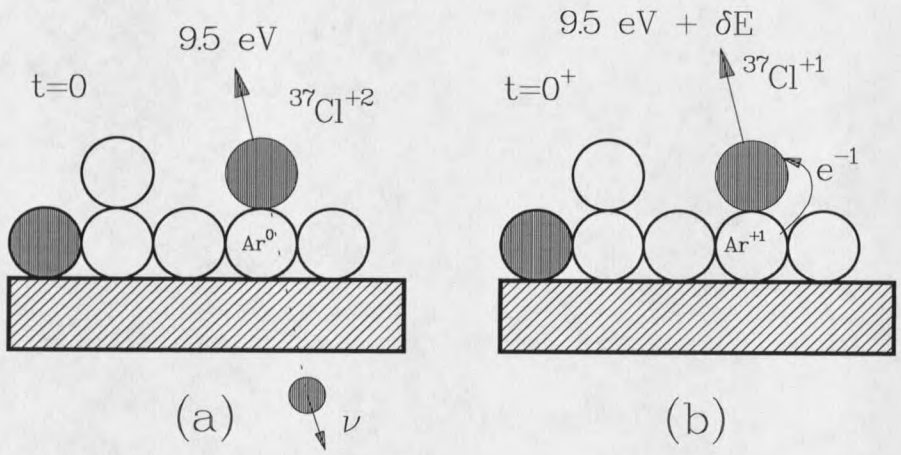

Figure 1.2 (a) ^{37}Cl ion obtains 9.5 eV recoil energy from neutrino emission. (b) Charge is transferred and Coulomb repulsion gives additional energy δE to the outgoing ^{37}Cl ion.

The second interaction to be discussed is the charge exchange between the ^{37}Cl ions and their local surrounding atoms.[17-21] Theoretical calculations and experiments have shown that in the gas phase the EC decay of ^{37}Ar will give rise to dominantly $^{37}Cl^{3+}$, the positively threefold-charged ions.[22, 23] It is energetically more favorable for the $^{37}Cl^{3+}$ ion to pick up an electron from the neighboring particle, such as an Ar atom, during the initial phase of desorption.[24] These charge exchanges, however, could significantly broaden the

recoil energy spectrum. The positively charged ^{37}Cl ions and their positively charged neighboring atoms will interact via the Coulomb repulsion giving additional contributions to the kinetic energy of the outgoing ^{37}Cl ions (Fig. 1.2). In some cases, the additional energy due to this "Coulomb explosion"[25] could reach as high as 6 eV. The charge exchange process and Coulomb repulsion are the main causes of the broadening of the ^{37}Cl recoil energy spectrum.

The charge exchange process can also cause an outgoing ^{37}Cl ion to lose all of its positive charges and become a neutral atom while desorbing from the surface.[26-28] The experimental setup in this work can only detect charged particles. An estimation of the neutralization cross section and ion desorption yield per electron capture decay will be given from the experimental and computer simulation results.

There are other interactions that affect the energy distribution of the ^{37}Cl ions desorbing from the substrate. Two examples of them are the thermal vibration of the ^{37}Ar atoms on the substrate and the recoil of the ^{37}Cl ions due to Auger electron emission. The physical and chemical characteristics of the substrates can also play an important role in affecting the energy distribution of the ^{37}Cl ions. Some of these effects will be calculated using computer simulation to compare with the experimental results.

A full treatment of each of the interactions discussed above is beyond the scope of this work, rather, the emphasis is put on the study of the "Coulomb explosion"

phenomenon which is of great significance for the understanding of this particular desorption process. Because this interaction is the dominant factor in the broadening of the recoil energy spectrum, it also becomes a major obstacle to the neutrino mass limit measurements. The broadening of the recoil spectrum reduces the sensitivity of the neutrino mass limit measurements in this experiment. However, through the present studies the major factors which are responsible for the broadening of the recoil spectrum are understood. Future experiments will be designed with reduced charge exchange effects.

Auger Relaxation of ^{37}Cl atom

The electron capture decay of the ^{37}Ar atom creates a highly unusual electronic configuration in the daughter ^{37}Cl atom which is not encountered in ordinary non-nuclear physical process. Conventional excitations utilizing electron or photon bombardments generally leave the atom in an ionized initial state. However, in the EC decay a proton and an electron become a neutron, hence the total net charge of the atom is unchanged. The daughter ^{37}Cl atom remains neutral, even though there is a vacancy in its inner shell —the hole resulting from the capture of a electron. As shown in Fig. 1.3, this initial state is unusual in three ways. First, the electronic configuration of the initial state in ^{37}Cl atom is like Cl in energy but resembles Ar in structure, *i.e.,* each energy level of the ^{37}Cl atom is relaxed from the Ar level to Cl level within the decay time but all levels except 1s are still fully occupied with electrons, as in the Ar case. Second, this initial state is a neutral