RESONANT COHERENT EXCITATION OF FAST HEAVY IONS IN CRYSTALS[†]

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ABSTRACT

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Hydrogen-like ions of B, C, N, O and F and helium-like ions of N, O and F have been shown to exhibit resonant coherent excitation from their $n = 1$ to their $n = 2$ states, caused by periodic electric potential oscillations which occur as the ion moves through axial or planar channels of a thin crystal of Au. The resonance velocities of the ions exhibit shifts and splittings which are the result of the crystal fields acting upon the one-, or two-electron ions. Comparisons with theory show fairly good agreement except for certain weak resonance components which do not appear or appear at unexpected values of the resonance velocity.

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1. Introduction

It is well-known that particles moving in crystal channels lose less energy than they would lose if moving in a random direction in the same crystal¹). The particles are repelled from regions of high electron density (and high stopping power) by correlated very small angle scatterings; the energy loss can be less by factors $\sim 1/2$. An additional fact which is very important for this work is that channeled particles tend not to change their charge states. In particular, one-electron oxygen ions with 27.8 MeV energy have been observed to travel distances $\sim 10,000\text{\AA}$ in crystal channels of Ag without a single change of charge state²). It was found that this stability of charge state holds for both planar and axial channeling but if the channeled particles have large excursions about the axis (or mid-plane) of a channel they will change charge frequently. "Best channeled" particles have greatest charge stability and these typically can be identified by the fact that they also have the least energy loss. The conditions for resonant coherent excitation are shown in fig. 1. As the ions pass along the channel they encounter electric fields which are spatially periodic. This spatial periodicity translates to a time periodicity which depends on the velocity of the ion ($\nu = v/d$ where d is the atom string spacing). The fields are not pure sine waves but they are composed of the fundamental frequency plus several harmonics of the string atom frequency ($\nu = K v/d$; $K = 1, 2, 3 \dots$).

The excitation energy required to excite the $n = 2$ state in a hydrogenic ion is represented by $(\Delta E)_{1,2} = h\nu_{1,2}$. If one of the harmonics of the crystal field coincides with $\nu_{1,2}$, resonance coherent excitation can take place. Illustrative examples are given in the figure. It is expected then that the crystal frequency can be tuned by varying the velocity of the ion,

and, at resonance, the crystal field would cause part of the ions to be in their first excited state.

Excited ions are expected to be more easily ionized since the binding energy of the electron is reduced and the "size" of the ion is larger than that of a ground state ion. Resonance can then be detected by a somewhat depressed population of one-electron ions (or enhanced population of bare ions) among the emerging particles.

The oscillating electric fields fall in two different classifications, viz. those which are transverse and those which are parallel to the particle trajectory. For the $\langle 100 \rangle$ axis illustrated in fig. 1 transverse fields for particles along the channel center line are zero for all harmonics. Longitudinal fields are non-zero only for even harmonics of the fundamental string frequency³). Differences between transverse and longitudinal fields will be discussed later.

2. Experimental Arrangement

To detect variations in the populations of various charge states the apparatus shown in fig. 2 was used. Ion beams of B, C, N, O and F were obtained from the Oak Ridge EN Tandem Accelerator. A gas stripper between the two magnetic analyzers was used to provide and select one-electron and two-electron ions. The beam to the crystal was collimated by two 0.5 mm apertures 1.5m apart and a gold crystal 850Å thick was used. The emerging channeling pattern was further collimated to select well channeled particles and these were passed into a magnetic spectrometer where two (and later three) position sensitive detectors provided charge state separation and energy loss analysis. Bare nuclei and one-electron ions were collected

simultaneously so that count rate ratios would be independent of ion beam current. As shown in fig. 3, the resonance condition causes a depression in the high energy (low loss) part of the one-electron population with a corresponding enhancement of the bare nucleus population. The ratio R is the total one-electron ion count divided by the sum of the one-electron and bare nucleus counts, with no spectrum stripping. Spectrum stripping has been found to be unnecessary except for a few very weak resonances. Typical resonance data are shown in fig. 4. Higher harmonics are weaker because of the effects of crystalline thermal motion. In many cases the resonances are shifted below the velocity which was calculated on the basis of the hydrogenic atom in free space. In our case, the hydrogenic atom exists in a crystalline medium when the electron density is not zero and the electric fields can be quite strong. As shown in fig. 5 some resonances consist of a double minimum; these occur for the $\langle 100 \rangle$ channel when the harmonic is even and the fields are predominantly longitudinal.

3. Nature of the Resonances

Longitudinal oscillating fields are parallel to the direction of the braking field in the crystal. Crawford and Ritchie have analyzed the effect of the various crystal fields upon the $N = 2$ state manifold of the hydrogenic atom⁴), and these effects are illustrated in fig. 6. The potential acting upon the ion is composed of one part called the static potential which is due to the crystal itself and one part called the polarization potential which is due to the motion of the ion. The static, time invariant part of the field breaks up the $N = 2$ state into a $2p_z$, a $2p_x-2p_y$, and a $2s$ state as shown in the center part of fig. 6. The polarization

field is caused by the fact that the electrons of the crystal are formed into a wake following the ion thus producing a braking field⁵). The polarization field is directed backward along the trajectory; the overall effect of both potentials is to produce Stark mixing of the $2s$ state and the $2p_z$ state. The energy levels are as shown on the right in fig. 6; the $2p_x$ and $2p_y$ components lie at the same energy and the $2sp_z^*$ level lies above the original degenerate level position. Oscillating fields which are transverse to the particle trajectory would excite only the $2p_x$, $2p_y$ components and give a single resonance minimum, and longitudinal oscillating fields would excite only the $2sp_z^*$ and $2sp_z$ components giving rise to a double resonance. In the actual case of our experiments where ions have all possible channeling trajectories, the transverse and longitudinal fields are always mixed. As an approximate rule for the $\langle 100 \rangle$ axis, the even harmonics tend to produce mostly longitudinal ($2sp_z^*$ and $2sp_z$) excitation and for odd harmonics mostly transverse (the single $2p_x$, $2p_y$ resonance) excitation.

4. Results

Comparisons of our data to theory are shown in figs. 7, 8 and 9. Theory is shown as open circles and experimental results are shown as full circles. The third harmonic cases in figs. 7 and 9 show excellent agreement between theory and experiment. The second harmonic cases in figs. 7 and 8 show good agreement for the upper $2sp_z^*$ state but very poor agreement for the lower $2sp_z$ state. A small admixture of transverse field in the second harmonic cases could produce $2p_x$, $2p_y$ minima which might obscure

the $2sp_z$ component; this difficulty has not been resolved and it persists in almost every case which we have studied.

5. Planar Channels

The influences of strings of atoms do not disappear for the case of planar channeling⁶). As shown in fig. 10, the ion passes near successive strings as it drifts to the right. In addition, the ion is influenced by strings along other directions and thus two frequencies operate simultaneously. For example, the expression in fig. 1 becomes, for the (100) planar channel:

$$E_T(\text{MeV/amu}) = 3.03 (\ell \cos\theta + k \sin\theta)^{-2} d_A^2 (\Delta E_{\text{keV}})^2. \quad (1)$$

A map of resonance energy versus angle in the (100) plane is shown as the dashed curves in fig. 11, for one-electron ions of nitrogen. The solid curves are drawn for transitions to the first excited state of two-electron He-like nitrogen. The two sets of curves are identical except for a scale factor which reflects the fact that $(\Delta E_{12})_H > (\Delta E_{12})_{He}$. The data shown in fig. 12 verify that the resonances occur at approximately the angles and energies predicted in fig. 11.

6. The Effect of Pathlength

For the case of channeling between (110) planes, the resonance energies are

$$E_T(\text{MeV/amu}) = 3.03 (\ell \cos\theta + 2^{-\frac{1}{2}}m \sin\theta)^{-2} d_A^2 (\Delta E_{\text{keV}})^2 \quad (2)$$

where ℓ is any positive integer and m is any even integer. The resonance map for one-electron oxygen is given in fig. 13. The crystal used in these experiments had its $\langle 111 \rangle$ axis normal to the crystal surface; thus tilting the crystal toward the $\langle 100 \rangle$ axis along the (110) plane produces an increase in pathlength.

With planar channeling it is possible to reach two angles which produce identical crystal geometry and resonance energies but with different pathlengths. The data shown in figs. 14 and 15 were recorded 10.3° from the $\langle 100 \rangle$ axis in the (100) plane on the side toward and away from the $\langle 111 \rangle$ axis, respectively. The thin side pathlength was only 59% of the thick side pathlength. The data show that additional pathlength has not caused deeper minima. The broadening which we observe may be caused by the fact that energy loss in the crystal brings the particles out of the resonance velocity range. The pathlength on the thin side was $1190\text{\AA}$ and this may be excessive for precise determinations of resonance profiles. Resonances for higher Z , higher velocity particles would carry the advantage that the percentage energy loss in the crystal would be greatly reduced.

7. Secondary Effects

Although many well-channeled particles will retain charge state stability, it is also true that, even for very thin crystals, some of these ions will change charge. A pure beam of one charge state will thus provide well-channeled particles of other charge states even in the absence of resonances; thus it is expected that helium-like and hydrogen-like resonances will occur together in the crystal since particles of both kinds will be available. Helium-like resonances will depress the two-electron population and enhance the one-electron population. In addition, the resonance will indirectly enhance the bare ion population which is fed from the one-electron population. Conversely, hydrogenic resonances will depress both the one-electron population and the two-electron population. These secondary effects can be determined by changing the charge state of the beam particles. An illustration of secondary effects is shown in fig. 16.

A hydrogenic double minimum near 44 MeV causes a depression in the (7+) population and a secondary depression in the (6+) population.

8. Planar Tipping

A variation of the mixture of channeling trajectories can be accomplished by a procedure called "planar tipping". If the planes are misaligned with respect to the beam, then particles which have the smallest transverse excursions about the mid-plane will be eliminated until, at large tilt angles, particles which can be coherently excited will no longer appear. The resonance extinction with tilt angle can be seen by comparing figs. 16, 17 and 18. The expectation had been that different parts of the resonance curve could be attributed to particles of different transverse excursion amplitude. These results seem to indicate that all parts of the resonance attenuate together, contradicting the original expectation. Marks have been provided on the curves of figs. 16-18 to show where hydrogenic (H) and helium-like (He) resonances would be expected and the planar channel harmonic indices are given.

9. Two Electron Resonances

Similar data have been taken for helium-like resonances. In this case, there is no degeneracy in 2s and 2p states of He and no Stark-mixed doublets are expected for the even-harmonic resonances. In fig. 19 we show both hydrogenic and helium-like resonances; here we have used two-electron O^{6+} input ions. Near 33 MeV, a minimum in the O^{6+} population is accompanied by a maximum in the O^{7+} population and a secondary maximum in the O^{8+} population is the indirect result of the increase in the O^{7+} population. Near 38 MeV a helium-like resonance causes a depression for O^{6+} and an enhancement of O^{7+}

but part of the O^{7+} enhancement is affected by a hydrogenic resonance which depletes O^{7+} and enhances O^{8+} . In this case, the hydrogenic resonance is formed entirely from ions which have already undergone one change of charge. With planar tipping as shown in figs. 20 and 21, the resonances extinguish uniformly without significant change of shape or centroid shift. The conclusions on resonance extinction must be regarded as preliminary. A complete set of data must include the use of input ions having zero, one and two electrons in order to evaluate and separate the primary and secondary maxima and minima.

10. Summary

We have found hydrogenic resonances for B, C, N, O and F ions and helium-like resonances for N, O and F ions. Both axial and planar resonances have been observed. Theoretical evaluations of the level shifts and Stark-splittings give generally good agreement with the experimental results with a possible exception that the lower member of the $2sp_z - 2sp_z^*$ doublet either has an energy discrepancy or is obscured by an admixture of the $2p_x 2p_y$ component. In planar channels hydrogenic resonances have been found with two-electron input ions indicating that ions can change charge and then undergo resonant excitation in the crystal. Crystal pathlength changes have been used to show that resonances occur in exceedingly short distances and that further pathlength increases result in energy losses which bring the particle energies below resonance. Preliminary data with crystal tipping indicate a uniform extinction of resonances and exhibiting no selectivity of different excursion amplitudes for different parts of resonance profiles.

References

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Figure Captions

1. Conditions for resonant coherent excitation of channeled ions.
2. Experimental arrangement.
3. Energy loss spectra of N^{6+} and N^{7+} ions collected below, at, and above resonance.
4. Ratio of N^{6+} counts to the sum of N^{6+} and N^{7+} counts as a function of initial energy for $\langle 111 \rangle$ axial channeling.
5. Resonance with double minimum.
6. Effect of crystal fields upon the $N = 2$ state of the hydrogenic ion in gold.
7. Comparison of theory with experiment.
8. Comparison of theory with experiment. For data points, see fig. 5.
9. Comparison of theory with experiment.
10. Trajectory in planar channeling with two planes, one above and one below the plane of the figure.
11. Resonance energies for the (100) planar channel in gold. The dashed lines are for hydrogenic nitrogen and the solid lines are for helium-like nitrogen.
12. Ratio $R = N^{6+} / (N^{6+} + N^{7+})$ in (100) planar channeling at $\theta = 6^\circ$ and 38° .
13. Resonance energies for hydrogenic oxygen in the (110) planes of gold.
14. (110) planar resonances of O^{7+} in gold. Pathlength 1190 Å.
10.3° from $\langle 100 \rangle$.
15. (110) planar resonances of O^{7+} in gold. Pathlength 2014 Å.
10.3° from $\langle 100 \rangle$.
16. Oxygen 7+ ions on Au 38° from 100 axis in 100 plane (Tilt Angle = 0°).

Figure Captions (contd)

17. Oxygen 7+ ions on Au 38° from 100 axis in 100 plane (Tilt Angle = 0.096°).
18. Oxygen 7+ ions on Au 38° from 100 axis in 100 plane (Tilt Angle = 0.146°).
19. Oxygen 6+ ions on Au 38° from 100 axis in 100 plane (Tilt Angle = 0°).
20. Oxygen 6+ ions on Au 38° from 100 axis in 100 plane (Tilt Angle = $.096^\circ$).
21. Oxygen 6+ ions on Au 38° from 100 axis in 100 plane (Tilt Angle = 0.146°).

FOR RESONANCE

$$h\nu = (\Delta E)_{ij} = hK v/d$$

FOR $i = 1, j = 2$

$$E_r(\text{MeV/amu}) = 3.03 K^{-2} d_{\text{\AA}}^2 (\Delta E_{\text{keV}})^2$$

$$E_r(\text{MeV/amu}) = 3.15 \times 10^{-4} K^{-2} d_{\text{\AA}}^2 Z_1^4$$

EXAMPLE: C^{5+} $\langle 100 \rangle$ AXIS IN Au

$d = 4.079 \text{ \AA}$ AT 25°C

$K = 1$ $E_r = 6.8 \text{ MeV/amu}$ (81.6 MeV)

$K = 2$ 20.4 MeV

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

COMPARISON OF THEORY WITH EXPERIMENT

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Fig. 7

Fig. 8

Fig. 9

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Fig. 10

Fig. 11

Fig. 12

Fig. 13

Fig. 14

Fig. 15

Oxygen 7+ Ions on Au 38° From 100 Axis in 100 Plane (Tilt Angle = 0°)

Fig. 16

Oxygen 7+ Ions on Au 38° From 100 Axis in 100 Plane (Tilt Angle = 0.096°)

Fig. 17

Oxygen 7+ Ions on Au 38° From 100 Axis in 100 Plane (Tilt Angle = 0.146°)

Fig. 18

Oxygen 6+ Ions on Au 38° From 100 Axis in 100 Plane (Tilt Angle = 0°)

Fig. 19

Oxygen $6+$ Ions on Au 38° From 100 Axis in 100 Plane (Tilt Angle = 0.096°)

Fig. 20

Oxygen 6+ Ions on Au 38° From 100 Axis in 100 Plane (Tilt Angle = 0.146°)

Fig. 21