

(2) Uncertainties in the nuclear wave functions. More recent Cabibbo-model predictions⁹ of $P_\gamma^{CC}(1.08 \text{ MeV})$ are in qualitative agreement with, and somewhat larger than, the results of Ref. 8.

(3) The "enhancement factor" to be expected from the neutral weak currents is difficult to calculate precisely; current estimates¹⁰ for the Weinberg-Salam model point to a value of about 10.

The ^{18}F system is one of the most favorable cases, both experimentally and theoretically, for studying $\Delta T = 1$ PNC forces. With the Cabibbo-model prediction of Ref. 8, the present experiment yields an upper limit of 7.5 for the "enhancement factor" produced by neutral weak currents.¹¹

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Resonant Coherent Excitation of Channeled Ions

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We have observed resonant excitation of swift channeled hydrogenlike ions ($Z = 5$ to $Z = 9$) and heliumlike F^{7+} which arises from a coherent periodic perturbation by the atoms in the bounding crystal rows. The resonance excitation was seen through the reduction in the transmission of fixed-charge-state ions through channels in thin crystals of Au and Ag.

A channeled ion passing between ordered rows of atoms in a crystal lattice with a velocity v experiences a coherent periodic perturbation of frequencies $\nu = k(v/d)$, $k = 1, 2, 3, \dots$, where d is distance between atoms in a row. When one of these frequencies coincides with $\nu_r = \Delta E_{ij}/h$, where

ΔE_{ij} is the energy difference between electronic states i and j of the ion, a resonant coherent excitation might occur. This possibility was first pointed out by Okorokov¹ but it requires well-defined states and perhaps this is the reason that previous investigations have yielded negative or,

at best, contradictory results. The four published experimental results were concerned with excitation of He^+ from the $n=1$ to the $n=4$ state and the radiation from $n=4$ to $n=3$ following emergence of the ion from the crystal. In the first of these papers Okorokov *et al.*² reported $\sim 10\%$ enhancement in this radiation for He^+ ions emerging from a $\langle 100 \rangle$ axial channel in a $\sim 1000\text{-}\text{\AA}$ Ag crystal. Gaillard *et al.*³ reported an even larger effect for the $\langle 110 \rangle$ channel in Au. However, recent definitive experiments by Berry *et al.*⁴, by Gaillard *et al.*⁴, and by Mannami *et al.*⁵ have shown no evidence for this resonance. The principal difficulty in visualizing the formation of a $\text{He}^+ n=4$ ion within the crystal is its "size" ($\sim 8 \text{\AA}$ in diameter). One might expect enhanced ionization, but emergent $\text{He}^+/\text{He}^{++}$ ratios showed a similar lack of resonant effect.⁴ In their theoretical treatment of the He^+ problem Shindo and Ohtsuki⁶ predict resonance peaks so broad that overlap between harmonics would totally obscure any coherent excitation.

We have utilized our previous observations that for ions of $Z=5$ through 9 ($E \gtrsim 1.5 \text{ MeV/amu}$) an appreciable fraction of zero-, one-, and two-electron systems can pass through the central channel cores ($\lesssim 1 \mu\text{m}$ thick) without charge change and that ions maintaining their input charge state can be identified by a characteristic energy loss peak.^{7,8} The probability of an ion remaining in a fixed charge state depends on the integrated flux of electrons encountered, and, very sensitively, on the state of the attached electron. For example, if an electron bound to a channeled ion makes an $n=1$ to $n=2$ transition, we should observe an increased probability for electron loss. We therefore measured the surviving fraction of ions in fixed charge states for a number of ionic species passing through channels in Ag (1000 \AA thick) and Au (850 \AA thick) as a function of velocity.

Ion beams obtained from the Oak Ridge National Laboratory tandem accelerator were passed through thin oriented single crystals. Emergent ions were collimated by a 1-mm aperture 1.5 m downstream and entered a magnetic spectrograph fitted with two position-sensitive detectors in the focal plane positioned to record the intensity and energy distributions for zero- and one-electron ions (e.g., $\text{N}^{6+}, \text{N}^{7+}$).

We have observed resonant excitation of $n=1$ to $n=2$ transitions in the H-like systems of B^{4+} in $\text{Ag}\langle 100 \rangle$ ($k=2$); C^{5+} in $\text{Ag}\langle 100 \rangle$ ($k=2$) and in $\text{Au}\langle 100 \rangle$ ($k=2$), $\langle 110 \rangle$ ($k=2$), $\langle 111 \rangle$ ($k=3, 4$), $\langle 114 \rangle$ ($k=6$); N^{6+} in $\text{Au}\langle 100 \rangle$ ($k=2, 3$), $\langle 110 \rangle$ ($k=2$), $\langle 111 \rangle$

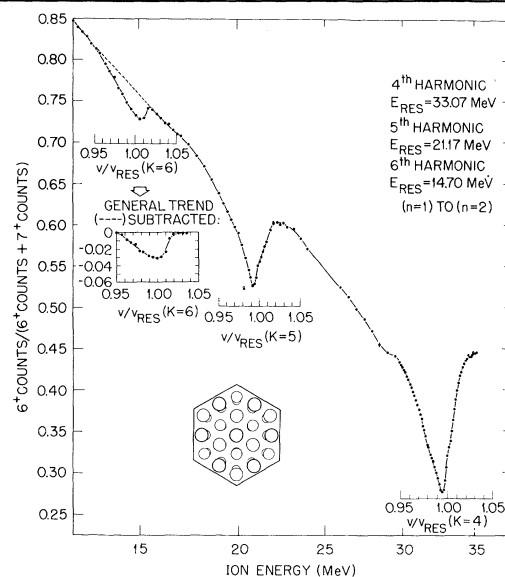


FIG. 1. Ratio of N^{6+} counts to the sum of N^{6+} and N^{7+} counts, as a function of initial energy, for $\langle 111 \rangle$ axial channeling in Au. The abscissa is linear in velocity. The incident beam is N^{6+} .

($k=4, 5, 6$); O^{7+} in $\text{Au}\langle 110 \rangle$ ($k=3$), $\langle 110 \rangle$ ($k=2$), $\langle 114 \rangle$ ($k=6$); and F^{8+} in $\text{Au}\langle 100 \rangle$ ($k=4$). The $n=1$ to $n=2$ transition for He-like F^{7+} in $\text{Au}\langle 100 \rangle$ ($k=4$) has also been observed. In some cases weak $n=1$ to $n=3$ transitions were observed. No $n=1$ to $n \geq 4$ transitions were detected.

We report here only those cases necessary to prove that the effect exists and to demonstrate some of its consequences. In Fig. 1 for incident N^{6+} we display the total population fraction of N^{6+} emerging from an 850- \AA -thick Au crystal as a function of the incident energy. The spacing along a $\langle 111 \rangle$ row is 7.064 \AA . Resonances are shown for $k=4, 5$, and 6. The additional scales shown are given in terms of v/v_r where v_r is the calculated resonant velocity.

Although the total $(Z-1)^+$ fraction is plotted in Figs. 1–3, the energy loss spectra for one-electron ions consist of two parts⁹: (1) fixed-charge-state ions which appear as a low-energy loss peak, and (2) partially charge equilibrated ions which have passed closer to atom rows and appear as a separable peak towards higher energy losses. By deconvolution we can show that the resonance affects *only* the fixed-charge peak. Hence, the actual dips are deeper than shown. However, for $Z=7$ to 9, the high-loss group is quite small, amounting to only, e.g., $\sim 15\%$ of the N^{6+} peak in the nonresonant regions of Fig. 1.

Consider first the processes occurring in the

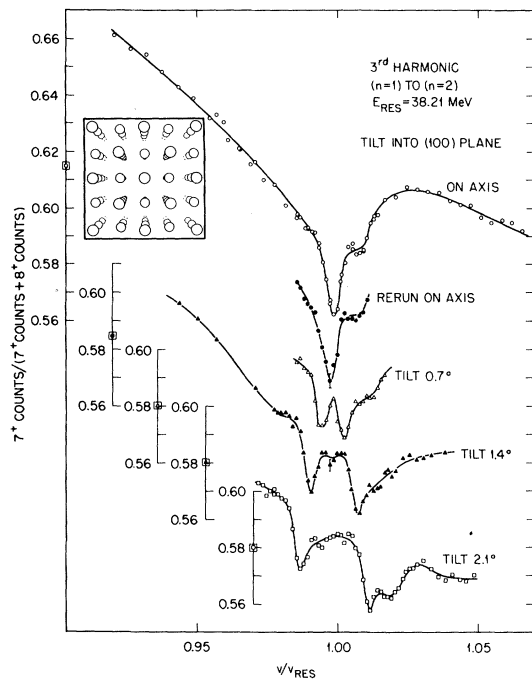


FIG. 2. Ratio of O^{7+} counts to the sum of O^{7+} and O^{8+} counts as a function of initial velocity, for $\langle 100 \rangle$ axial channeling in Au and for three tilt angles. The incident beam is O^{7+} .

absence of resonant excitation. For example, 35-MeV N^{6+} ions passing on a well-channeled course through $\sim 1000 \text{ \AA}$ of Au will encounter an electron target thickness of $\sim 5 \times 10^{18} / \text{cm}^2$ (~ 10 electrons/Au atom) and the center-of-mass energy is 1400 eV. By electron collision,⁹ the ion can be directly ionized ($\sigma_{1s}^i \sim 5 \times 10^{-20} \text{ cm}^2$) or excited to $n=2$ ($\sigma_{1s-2p} \sim 5 \times 10^{-20} \text{ cm}^2$) and then ionized ($\sigma_{2p}^i \sim 5 \times 10^{-19} \text{ cm}^2$). Once ionized, the probability of recapturing an electron is small^{6,7}; the passage would leave $\sim 50\%$ of the N^{6+} ions unchanged, in rough accord with the observations. The coherent resonant excitation process to $n=2$ effectively supplies an additional ionization channel.

For all the simpler features of the resonances, some generalizations can be made: (1) Narrower channels give stronger resonances, (2) higher harmonic resonances are weaker, (3) there are slight shifts in the resonance peaks to lower (v/v_r), and (4) the peaks are asymmetric towards lower (v/v_r). The first is easily understood, since ions moving through narrower channels experience stronger perturbations. The remaining three can all be seen in Fig. 1. The peak shifts may be caused by the decreased $n=1$ to $n=2$ energy spacing due to the presence of electrons

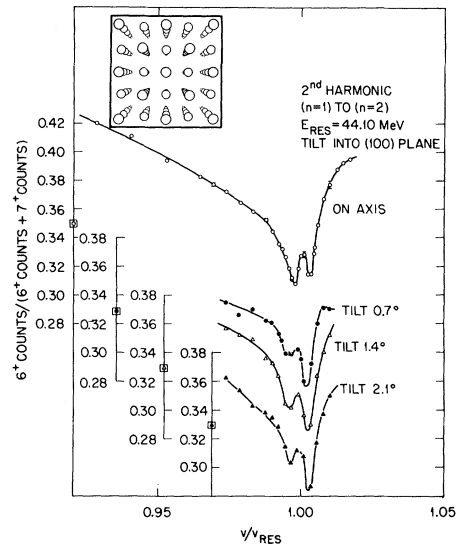


FIG. 3. Ratio of N^{6+} counts to the sum of N^{6+} and N^{7+} counts as a function of initial velocity, for $\langle 100 \rangle$ axial channeling in Au and for three tilt angles. The incident beam is N^{6+} .

from the medium contained within the $n=1$ and $n=2$ shell radii. The spreading is asymmetric since the minimum density is at the exact channel center and increases as the rows are approached.

Some more subtle features of the resonances are shown in Figs. 2 and 3 for the $\langle 100 \rangle$ channel ($d = 4.078 \text{ \AA} = a$, the lattice constant). In Fig. 2 we show a third (odd) harmonic excitation for O^{7+} and the splitting of the single central feature as the crystal axis is tilted in the $\langle 100 \rangle$ planar direction. In Fig. 3 we show a double peak seen for the second harmonic of N^{6+} directly on axis, and its insensitivity to tilt angle. Evidence for doublet structure has been seen for all ions in *even* harmonics in the $\langle 100 \rangle$ and $\langle 110 \rangle$ axes. We will show that the splitting in Fig. 2 is due to sidebands produced by modulation of the central frequency when the crystal is tilted, while the splitting in Fig. 3 is due to Stark splitting of the $n=2$ levels of the ion.

An ion channeled along the $\langle 100 \rangle$ axis (z direction) generates an electron wake.¹⁰ This produces a field which can be shown to split the $n=2$ manifold of the ion into three levels, the central one of which is $2p_x, 2p_y$ (degenerate), while the outer ones arise from mixed $2s, 2p_z$ states. We attribute the central minimum in Fig. 2 to transitions to the degenerate $2p_x$ and $2p_y$ states, and the two large minima in Fig. 3 to transitions from $1s$ to the mixed $2s, 2p_z$ levels.

Selection rules have been derived from the electric potential, which in an unperturbed face-centered-cubic crystal can be shown to have the following form:

$$V = \sum_{klm \text{ even}} V_{klm} \cos(2\pi kz/a) \cos(2\pi lx/a) \cos(2\pi my/a) + \sum_{klm \text{ odd}} V_{klm} \sin(2\pi kz/a) \sin(2\pi lx/a) \sin(2\pi my/a), \quad (1)$$

where the z , x , and y axes are centered in $\langle 100 \rangle$, $\langle 010 \rangle$, and $\langle 001 \rangle$ channels. The two sums are over all sets of nonnegative even and odd integers, respectively. The first sum contains the even harmonics, and the second sum the odd harmonics, of the fundamental frequency, v/a . One finds, for sufficiently small x and y , that the oscillating field is predominantly in the x and y directions for odd harmonics (odd k), and in the z direction for even harmonics. Dipole selection rules have been shown to apply. Thus, when the ion comes into resonance with an odd harmonic, as in Fig. 2, the strongest transitions should be those to the degenerate $2p_x$ and $2p_y$ states, while resonance with an even harmonic, as in Fig. 3, should favor the mixed $2s$, $2p_z$ states. It follows that the separation between the two minima in Fig. 3 is a measure of the wake field in the vicinity of the moving ion. If we assume a uniform field \mathcal{E} , the first-order Stark splitting is $6e\mathcal{E}a_0/Z$. The data of Fig. 3 give a value of $0.10e^2/a_0$ for this splitting, from which we deduce $\mathcal{E} = 0.12e/a_0^2$ which is of the expected order of magnitude.⁹ A more refined treatment is in progress.

If the crystal is rotated through an angle θ about the y axis, a similar analysis (assuming only small y) shows that, for even k , the field contains the following frequencies

$$(v/a)(k \cos\theta \pm l \sin\theta), \quad l = 0, 2, 4, \dots, \quad (2)$$

where the central ($l=0$) frequency component is mainly in the z direction. Therefore, for small tilts, one should find the same spectrum as for $\theta=0$, with the possible addition of small sidebands. Hence the curves in Fig. 3 show little change with tilt.

Odd harmonics behave differently; Fig. 2 shows a central minimum breaking into two minima. This occurs because the frequencies for odd k are as in Eq. (2), but here with only odd l . The ion steps through a succession of $\langle 100 \rangle$ channels. Since fields vary across a channel, an amplitude modulation results and sidebands appear. The central frequency is missing when k is odd because the strings of atoms which define the $\langle 100 \rangle$ channels in a face-centered-cubic crystal are staggered in z in such a way that the odd- k sig-

nals change phase by one cycle as the trajectory progresses sideways a distance a . A tilt of 0.7° causes the ion to step over a distance a once every 80 atoms of its passage (thus once per 240 cycles of the third harmonic), giving sidebands 0.4% above and below the frequency $k(v/a)$ ($\pm 0.4\%$ in v/v_r) in agreement with Eq. (2) with $l=1$. This argument contains the assumption that an ion "remembers" the phase of earlier signals. The splitting shown in Fig. 2 is direct evidence that here excitation occurs coherently over ~ 80 or more unit cells.

The existence of these effects requires that well defined states of the ion exist within the solid over considerable distances. Since ionization cross sections for H-like ions vary⁹ as Z^{-4} , electron loss from even the $1s$ state of He^+ is probably too rapid for appreciable coherent effects to exist¹⁻⁴; the absence of observable resonances to states $n > 3$ for ions of $Z \leq 9$ is in part due to the short collisional lifetimes of these states.

Ions with higher Z have higher binding energies and smaller shell radii; some excited states should survive passage through thin crystals and would then decay by x-ray emission. Our work on F^{8+} ions suggests that a large portion of the $n=2$ states emerge so that studies of x radiation and polarization may add significant information (e.g., odd and even harmonics might yield different $2s/2p$ fractions with differing polarization). With reduced probability of ionization, effects of downward resonant transitions, resonance excitation to $n > 3$ levels, and multiple resonance excitation may appear. Finally, for higher- Z ions, multiple-electron systems are available for study.

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High-Resolution Two-Photon Spectroscopy with Picosecond Light Pulses

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We have demonstrated the feasibility of Doppler-free two-photon spectroscopy with a train of picosecond standing-wave light pulses from a synchronously pumped mode-locked cw dye laser. The actively controlled mode spectrum provides a means for accurate measurements of large frequency intervals. From a multipulse spectrum of the sodium $3s$ - $4d$ transition we have determined a new value of the $4d$ fine-structure splitting, 1028 ± 0.4 MHz.

Two-photon excitation of gas atoms with a coherent train of standing-wave light pulses can produce fringelike spectra free of first-order Doppler broadening, which resemble the spatial diffraction pattern of a multislit grating or the Ramsey fringes of molecular beam spectroscopy with multiple spatially separated rf field regions.¹⁻⁵ Shortly after the possibility of Doppler-free two-photon spectroscopy with coherent pulse trains was first suggested,¹ narrow spectral fringes were observed by exciting the sodium $3s$ - $5s$ transition with nanosecond pulses recirculating inside a passive optical resonator.²

In this paper we report on experiments which establish the feasibility of high-resolution spectroscopy with a synchronously pumped actively mode-locked cw dye laser. We have recorded Doppler-free spectra of the sodium $3s$ - $4d$ transition with pulse ranging in length from 500 psec to less than 1 psec. The actively controlled frequency spacing of the phase-locked laser modes provides a precise frequency calibration scale. This makes it possible to apply accurate electronic frequency counting techniques to the measurement of large line separations, up to high multiples of the pulse repetition rate, where direct modulation or beat-frequency techniques would be difficult or impossible to apply. Using

this calibration, we have determined a new value of the sodium $4d$ fine-structure splitting, 1028.5 ± 0.4 MHz.

The origin of the narrow spectral fringes in multipulse excitation is perhaps most easily understood in the frequency domain.²⁻⁴ The pulse train from a mode-locked laser can be described as a superposition of phase-locked oscillating laser modes which are equally spaced in frequency.⁶ Similarly, a pulse circulating inside a passive optical resonator transforms into a comb of "shock-excited" cavity modes. Resonant two-photon excitation is possible whenever the sum of two mode frequencies coincides with an atomic transition frequency. Many pairs of modes can satisfy this resonance condition at the same time and contribute to the resonant excitation, and a detailed analysis must consider the spatial field distribution and phases of all these modes.^{3,4} If the atoms are in a region where two counterpropagating pulses form a standing-wave field, this excitation can be nearly free of first-order Doppler broadening. The signal magnitude is of the same order as under cw excitation with a single-frequency laser of the same average power.

The multipulse excitation can also be described in the time domain.^{1,2,5} Atoms which have been excited to a coherent superposition of ground