Isotope Separation in Plasmas by Use of Ion Cyclotron Resonance*

J. M. Dawson,† H. C. Kim, D. Arnush, B. D. Fried, R. W. Gould,‡ L. O. Heflinger, C. F. Kennel,† T. E. Romesser, R. L. Stenzel, A. Y. Wong,† and R. F. Wuerker
TRW Defense and Space Systems, Redondo Beach, California 90278
(Received 23 September 1976)

Selective ion-cyclotron-resonance acceleration of individual K, Ne, Cl, A, and Xe isotopes has been observed in plasmas of density 10^9 to 10^{11} cm^{-3}. Energy discriminating probes show good resolution of accelerated species in all cases. Mass spectrometer analyses of potassium samples, collected on cooled tungsten ribbons, showed ^{41}K to ^{39}K abundance ratios of 4 rather than the normal value of 0.07.

The use of ion cyclotron resonance for heating fusion plasmas is well known.[1] In applying this technique to separation of isotopes with a small fractional mass difference, $\Delta M/M$, we begin with a plasma immersed in a very uniform magnetic field ($\Delta B/B < M/M$) whose transverse dimensions exceed the maximum cyclotron radius of the isotopic species (mass M) to be accelerated. Applying an rf electric field with frequency equal to the cyclotron frequency, $\Omega = qB/Mc$, or a harmonic thereof, results in selective acceleration. Physical separation of that species, *throughout the plasma volume*, can then be effected by any of a wide variety of energy-dependent processes as will be discussed below. The physical separation of ^{41}K reported here employed the simplest of such processes, namely the collection of potassium on cold tungsten ribbons, immersed in the plasma and biased sufficiently positive to repel ions with energy below a few eV.

Selective excitation of one ionic species has previously been observed in hydrogenic (H^+, H_2^+) plasmas[2] and is contemplated in fusion applications (e.g., heating one component in a D, T plasma[3]), but resolution becomes much more difficult for heavy isotopes. An experiment by Schmitt,[4] carefully designed to produce "pure" ion Bernstein waves, did show a small jump in the dispersion curve arising from the ^{41}K component of a potassium plasma, but experiments by Richardson and others[5] with uranium compounds were unable even to approach the resolution required to separate uranium isotopes. Necessary conditions for the resolution of masses M and $M + \Delta M$ are (1) $\Omega\tau(\Delta M/M) > 1$, where $\tau = L/a_\parallel$ is a typical ion transit time (at parallel thermal velocity $a_\parallel$) along the length, L, of the magnetic field; and (2) $\nu/\Omega < \Delta M/M$, where ν is the average collision frequency of the energized ion species. Collision and transit time effects are not independent. While condition (2) quarantees adequate acceleration time for the resonant species, collisions still result in transfer of energy from motion transverse to B, to motion parallel to B, thus increasing the BL product required by condition (1). Collisions also transfer energy from the resonant species to nonresonant ones, spoiling the desired selective energization. Fortunately, the ion-runaway[6] phenomenon alleviates these difficulties, so that plasma densities as high as 10^{12} **or** 10^{13} cm^{-3} appear feasible.

The accelerating rf electric field may be produced *inductively*, by superimposing an ac component on the uniform magnetic field, as in magnetic pumping,[7] or *directly*, by applying an ac voltage between two electrodes located at one end of the plasma and biased so that they determine the plasma potential along the field lines which they intercept. Results using both methods are reported here. Inductive acceleration provides some control over the radial and axial wave numbers, $k_\perp$ and $k_\parallel$, of the exciting field. However, the large ac coil voltages used ($V_{rms} \simeq 1000$ V) require not only a high rf power level but also a slotted conducting cylindrical lining to shield the plasma from the axial rf fields. The necessity for having one or more axial slits leads to complicated rf field patterns. While direct excitation requires very little power (of order 3 W), the rf field in the plasma arises from the axial propagation of an ion cyclotron wave, whose properties are not easy to compute, particularly if one takes into account radial variations of plasma density, wave attenuation consequent on the resonant particle acceleration, etc.

Two different plasmas have been used: a small potassium Q machine[8] ($B \leq 4$ kG) with inductive excitation; and a large dc discharge plasma, immersed in a very uniform magnetic field ($\Delta B/B \leq 7 \times 10^{-3}$, $B \leq 20$ kG, produced by a superconducting coil) with direct excitation. (We have also used dc discharge sources to study other gases in the small machine, with both inductive and direct excitation, and have used inductive excitation in

the large device; the results are similar to those given here.)

The Q-machine plasma column is 5.7 cm in diameter and 1 m long. Typical plasma parameters are $n \simeq 10^{10}$ to 10^{11} cm^{-3}, $T_i \simeq T_e \simeq 0.2$ eV, $\nu_{i-i} \simeq 2\times10^4$ sec^{-1}, and $\nu_{e-i} \simeq 5\times10^6$ sec^{-1}. The rf pump field (80 to 100 kHz) is produced with a long solenoid (60 cm long, 15 cm diam, 47 turns) wound on a liquid-nitrogen-cooled aluminum cylinder with a single axial slot. The resultant azimuthal vacuum induction electric field is $E_{rms} = 0.06r$ (V/cm) for 300-W pump power, where r is the radial position in centimeters. The static B field is measured to an accuracy of 0.3%.

An energy-analyzing probe similar to the one used by Motley and Kawabe[9] is utilized to study the particle dynamics in the perpendicular direction. It has a platinum collector (0.2 cm diam) which is recessed (0.1 cm) in a cylindrical metallic shield so that thermal electrons cannot reach the collector ($r_{ce} \simeq 10^{-3}$ cm) while ions can ($r_{ci} \gtrsim 0.1$ cm). Resonant ions tend to bunch in phase space and enter the collector of the energy-analyzing probe as a burst, once each pump period. The cyclotron resonances are monitored by plotting the time-averaged collector current as a function of the static B field for a fixed pump frequency as in Fig. 1(a). The observed resonance frequencies deviate from the computed ion cyclotron frequencies by less than 1%, well within experimental errors. The resonance width decreases with increasing collector retarding voltage [Fig. 1(a)].

The variation of collector current with collector retarding potential is shown in Fig. 1(b). Both species are accelerated to energies of order 50 eV when driven at their resonant frequencies; at larger energies, r_{ci} would exceed the plasma column radius (2.8 cm), leading to large space-charge effects. The approximately exponential dependence of collector current indicates a Maxwellian distribution for the perpendicular ion energy. The higher "temperature" of the ^{41}K (15 eV vs 10 eV for ^{39}K) and the approximate equality of the maxima in the resonance curves, notwithstanding the thirteenfold predominance of the ^{39}K, is consistent with the fact that radial space charge for the species can be compensated only by axial electron motion, while for the minor species it can be balanced by small-amplitude, out-of-phase motion of the major species.[10]

Macroscopic samples of potassium were collected on a cooled (-5°C) tungsten ribbon, biased +4 V above the plasma potential so that it received only ions with energy exceeding 4 eV. After 30 minutes of collection, the plasma was turned off; the ribbon was brought to a quadrupole mass spectrometer mounted in the same vacuum system; and the sample was evaporated by ohmic heating of the tungsten ribbon. Since the potassium vapor was contact-ionized on a hot tungsten wire at the entrance of the mass spectrometer, other molecules with the same masses were not ionized. To eliminate spurious effects, samples were collected under three different conditions of rf excitation: no rf; excitation at the ^{39}K cyclotron frequency; and excitation at the ^{41}K cyclotron frequency. The resulting ratios of ^{41}K/^{39}K abundance as measured by the mass spectrometer are, respectively, 0.07 (the natural abundance), 0.02, and 4, the latter corresponding to a more than tenfold enrichment of ^{41}K. Typical mass spectrometer traces are shown in Fig. 2.

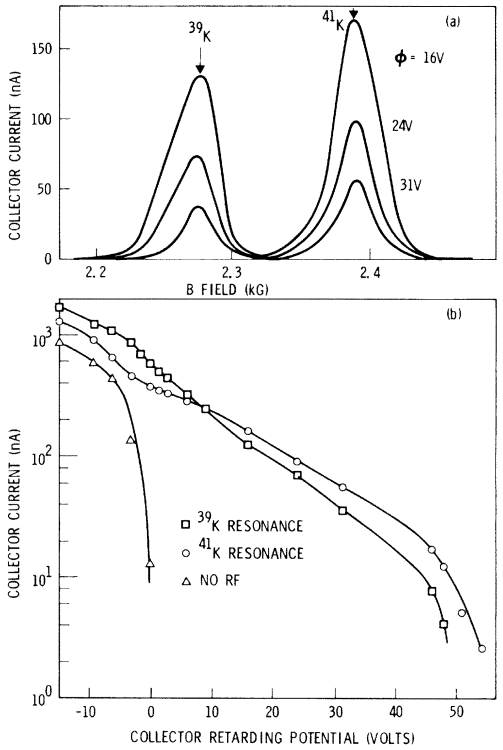

FIG. 1. (a) Ion cyclotron resonances of ^{39}K and ^{41}K measured by the radial energy analyzer. rf frequency = 88.6 kHz, rf power = 300 W, and φ is the retarding voltage. (b) Collector current vs collector retarding potential of the radial energy analyzer for ^{39}K and ^{41}K resonances. rf power = 300 W and rf frequency = 88.6 kHz.

For the selective acceleration of ions with mass greater than 60 the superconducting solenoid was used. One source used was a dispenser cathode

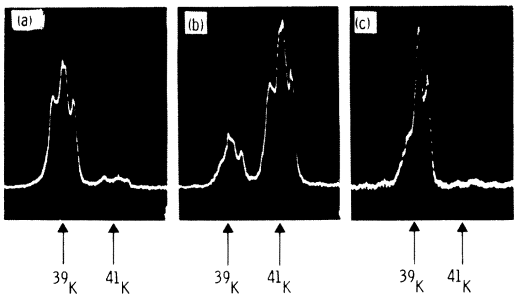

FIG. 2. Mass spectrum analysis of the potassium samples collected on a cold tungsten ribbon. Small fluctuations at the peak of each mass number are due to inherent noise in the mass spectrometer. The tungsten ribbon is biased +4 V with respect to the plasma. During the collection, the rf pump is (a) turned off, (b) tuned to ^{41}K, and (c) tuned to ^{39}K.

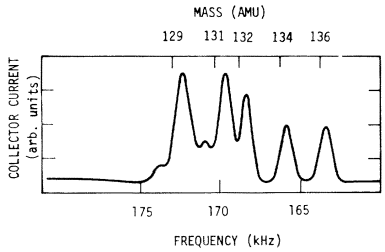

FIG. 3. Current detected by the radial energy analyzer as the applied rf frequency is swept at constant magnetic field. The magnetic field was measured to be 14 512 G. The mass values indicated at the top of the figure were calculated from the measured field.

discharge similar to the Stenzel Large Magnetized Plasma Source (LAMPS).[11] A 10×10-cm^2 dispenser cathode made of barium-oxide-impregnated tungsten was heated to emit electrons thermionically. Split anode plates were placed at the opposing end of the solenoid and served as the direct-excitation electrodes. A large variety of ions was studied. Xenon isotopes were investigated by using an argon discharge plasma ($p\sim10^{-4}$ Torr) to which a small quantity of xenon had been added ($p_{xe}\sim5\times10^{-6}$ Torr). Figure 3 shows a xenon resonance trace in which the radial energy analyzer current is plotted versus the rf frequency. The magnetic field was measured by a proton NMR. The mass scale labeled on the figure corresponds to the cyclotron frequencies derived from the NMR. The dominant xenon isotopes are $M=129$, 131, 132, 134, and 136; the absolute natural abundances are 26.4, 21.2, 26.9, 10.4, and 8.9%, respectively. The very small (less than 0.3%) uniform frequency shifts from the single-particle xenon ion cyclotron frequencies is attributable to collective effects which are currently being investigated. The relative amplitudes of the peaks are in good agreement with the natural abundance ratios, although at higher xenon concentrations and different operating conditions the observed ratios and frequencies deviate from these values.

In summary, we have observed well-resolved ion cyclotron resonances for the individual positive ions of isotopes of K, Ne, Ar, and Xe and the individual negative ions of the isotopes of Cl. We have also collected a macroscopic sample of potassium whose isotopic composition ratio as measured with a mass spectrometer is $[^{41}\mathrm{K}]/[^{39}\mathrm{K}]=4$. Resonance lines associated with more than thirty other atomic and molecular ions from hydrogen to uranium have been resolved, the minimum percentage linewidth being 0.4% full width at half-maximum. The plasma sources used in these experiments did not provide plasma densities above 10^{11} cm^{-3}, but similar sources have been operated at densities up to 10^{13} cm^{-3}. While collisional effects probably prevent operation at higher densities, this is still large enough to be of practical interest, particularly since this approach appears to be capable of single-pass, high enrichment. In addition to the use of cooled surfaces, physical separation techniques under study include: the use of magnetic mirrors to concentrate preferentially the energized species in the center of the plasma (an effect we have observed for ^{41}K$^+$ in experiments with weak mirrors)[12]; interaction of selectively energized molecular ions with either inert background molecules (spallation) or reactive molecules (endothermic chemical reactions) so that the energized species is brought to a different chemical form, permitting chemical separation; and differential diffusion across the magnetic field by the energized ions. Throughput can be enhanced by not only working at high density but also using $\vec{E}\times\vec{B}$ drifts to transport the plasma through the device at speeds exceeding the thermal velocity. If a given atomic species is present in more than one molecular form, several resonant frequencies may be applied simultaneously. Requirements on transit time and collision frequency can be relaxed by using excitation at cyclotron harmonics, since $k_\perp r_{ci}$ will be large for the energized species, but the efficiency will be reduced. Although the radial dependence of the rf fields leads to ion space charge, it is easily neutralized by axial motion of the electrons, in the case of a major species, and by out-of-phase radial motion of the major

species, in the case of the minor species. Thus, theory predicts resonance frequencies very close to the single-particle ion cyclotron frequencies and this is verified experimentally.

*This work supported by TRW Independent Research and Development and by U. S. Energy and Research Development Administration under Contract No. E(49-15)-3086L.

†Permanent address: University of California, Los Angeles, Calif. 90024.

‡Permanent address: California Institute of Technology, Pasadena, Calif. 91109.

[1]T. H. Stix, Nucl. Fusion 13, 737 (1965). This also gives many references to earlier work.

[2]H. Toyoma, J. Phys. Soc. Jpn. 34, 527 (1973); H. Toyama and K. Matsuura, J. Phys. Soc. Jpn. 35, 277 (1973).

[3]S. Yoshikawa, M. A. Ruthman, and R. M. Sinclair, Phys. Rev. Lett. 14, 214 (1965).

[4]J. P. M. Schmitt, Phys. Rev. Lett. 31, 982 (1973).

[5]J. R. Richardson, *National Nuclear Energy Series* Div. 111 (McGraw-Hill, New York, 1949), Chap. 12.

[6]H. Dreicer, Phys. Rev. 115, 238 (1959), and 117, 329 (1960).

[7]J. M. Berger, W. A. Newcomb, J. M. Dawson, F. A. Frieman, R. M. Kulsrud, and A. Lenard, Phys. Fluids 1, 301 (1958).

[8]R. W. Motley, *Q Machines*, (Academics, New York, 1975).

[9]R. W. Motley and T. Kawabe, Phys. Fluids 14, 1019 (1971).

[10]J. M. Dawson and B. D. Fried, to be published.

[11]R. L. Stenzel, Phys. Fluids 19, 857 (1976).

[12]H. C. Kim, to be published.

Ionic Conductivity near an Order-Disorder Transition: $RbAg_4I_5$†

R. Vargas,* M. B. Salamon, and C. P. Flynn

Department of Physics and Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

(Received 16 August 1976)

We report the first precise study of atomic mobility through a second-order phase transition. Simultaneous measurement of the ionic conductivity σ and the specific heat near the 208-K transition of $RbAg_4I_5$ establishes an accurate proportionality between $\ln\sigma$ and the ordering enthalpy h. This is interpreted using a many-body theory which treats local order beyond the mean-field approximation and in which the constant of proportionality is determined by microscopic details of the interaction among the mobile defects.

The mobile interstitials in some solids can diffuse with relative freedom among partially occupied sites connected by jump paths. In certain cases, the interactions among these mobile ions cause them to adopt partially ordered structures as the temperature is lowered.[1,2] Most treatments of the effect of ordering on the ionic mobility employ mean-field models which ignore important critical-point effects, and therefore predict that the activation energy should be unaffected by the phase transition above T_c and should follow the square of the long-range order parameter below.[3] An exception to this approach is Mahan's attempt[4] to treat a lattice-gas model rigorously. However, no accurate prediction for the ionic mobility near the phase transition was obtained. In this Letter we present the first experimental determination of atomic mobility near a phase transition sufficiently accurate[5,6] to examine the *critical* behavior, and the first theoretical analysis to go beyond the mean-field approximation.[3,7] We establish, both experimentally and theoretically, that an accurate proportionality exists between the logarithm of the ionic conductivity, $\ln\sigma$, and the interaction enthalpy, h, of the mobile-ion subsystem. This demonstrates the dominance of short-range order effects which have generally been ignored, and, through the constant of proportionality, gives microscopic information about the way short-range order modifies the energetics of the diffusive jump. The solid electrolyte $RbAg_4I_5$ has been employed because its conductivity provides a simple measure of mobility and because its 208-K phase transition has been shown to be Ising-like.[8]

The specific heat and the derivative dR/dT of the resistivity of $RbAg_4I_5$ single crystals were measured simultaneously as functions of temperature using the ac method.[9] Crystals were grown in our laboratories using the solution technique with high-purity reagents.[10] Small single crystals were chosen as representative specimens for crystallographic identification. Buerger precession patterns showed quite uniform alignment and a cubic unit cell with a lattice constant of 11.24 Å, in good agreement with previous determinations.[11] Chemical analysis yielded a Ag-Rb atomic ratio of 4.2 ± 0.2.

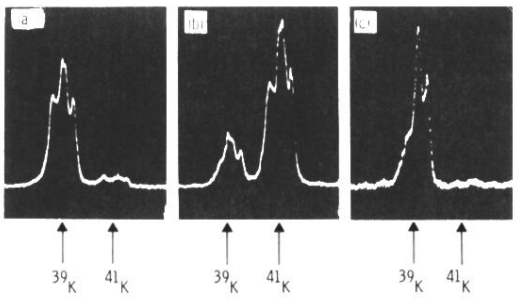

FIG. 2. Mass spectrum analysis of the potassium samples collected on a cold tungsten ribbon. Small fluctuations at the peak of each mass number are due to inherent noise in the mass spectrometer. The tungsten ribbon is biased +4 V with respect to the plasma. During the collection, the rf pump is (a) turned off, (b) tuned to ^{41}K, and (c) tuned to ^{39}K.