Optogalvanic isotope enrichment of Cu ions in Cu–Ne positive column discharges

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The isotopic enrichment of copper ions in a positive column Cu–Ne discharge using optogalvanic excitation is analyzed with a rate equation model. With excitation at 510.6 nm, the fraction of the ions belonging to the 63-amu isotope of copper is enriched relative to the neutral abundance. Enrichment as large as 10% is calculated when the initial abundance of the neutral isotope is small (≤ 0.1) and the discharge current density is large (≥ 75 mA/cm^2). The degree of enrichment is examined as a function of the initial abundance, discharge current, the rate of charge exchange, and the diameter of the discharge tube.

I. Introduction

The optogalvanic effect (OGE) is the perturbation of the current–voltage characteristic of a gas discharge by the absorption of radiation.[1,2] Typically a laser is tuned to a resonance or higher-lying transition in one of the components in the discharge. If absorption of the laser radiation increases the density of atoms in the state higher in energy, the stepwise ionization rate out of that state is increased. If the current through the discharge is held constant, the increase in electron density resulting from the larger ionization rate decreases the impedence of the discharge, thereby reducing the voltage drop between the electrodes. This change in voltage is the optogalvanic signal. By tuning the laser across a transition, the optogalvanic signal reproduces the absorption line shape. Using this technique, the OGE has found application as a spectroscopic tool.[3–6]

In an earlier work it was suggested that the OGE could be used as a method for isotope separation.[1] To enrich a particular isotope, a laser would be tuned to a transition in that isotope, preferentially ionizing it and increasing the fraction of ions of that isotope relative to the neutral abundance. An ion extraction technique would then be used to harvest the enriched ions.

Although the OGE has been observed in a variety of discharges, not all discharge types are suitable for isotopic enrichment by the OGE. Specifically, hollow cathodes appear to be unsuitable.[7,8] In a hollow cathode the bulk of ions is initially produced in the buffer gas by the e-beam component of the electron distribution which results from electrons emitted from the cathode.[9] Metal ions (the species of interest) are produced primarily by charge exchange with the buffer gas. This nonselective charge exchange reaction would likely overwhelm any enrichment gains made as a result of the OGE. A second factor which makes the prospect of isotope separation in a hollow cathode less than promising has to do with the details of the electron distribution. The region in the middle of a hollow cathode is almost free of any electric field, and as a result, the electron distribution is in equilibrium with the neutral gas. Superelastic relaxation of higher-lying levels excited by the laser radiation could add sufficient energy to the electron gas to increase the electron temperature.[7,8] The increase in the electron temperature would result in an increase in the nonselective electron impact ionization rate of ground-state isotopes or of isotopes excited to lower-lying states.

In a positive column discharge the restrictive effects discussed with respect to optogalvanic isotope enrichment in a hollow cathode do not apply. Under typical conditions the fraction of ions belonging to the buffer gas is negligible, thereby eliminating charge exchange collisions with the buffer gas as a source of nonselective ionization. The OGE in a positive column decreases the electron temperature for all conditions of practical interest rather than increasing the electron temperature as may be possible in a hollow cathode. Therefore, nonselective ionization of the ground state by electron impact is reduced instead of increased.

In this paper the isotopic enrichment of copper ions in a positive column Cu–Ne discharge under the conditions of quasi-steady-state optogalvanic excitation is

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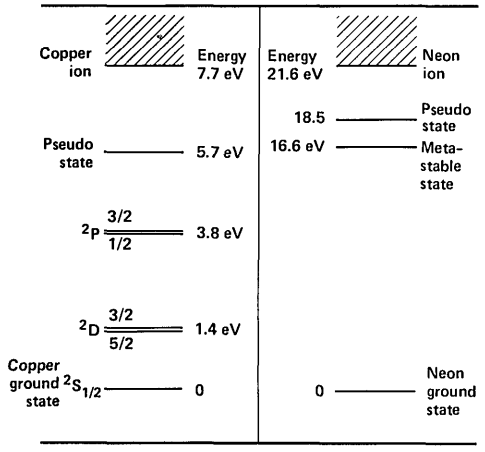

Fig. 1. Species included in this analysis. A set of copper levels is included for both copper isotopes. The stimulating radiation at 510.6 nm is between the $^2D_{5/2}$ and $^2P_{3/2}$ states.

analyzed with results from a rate equation model. This analysis differs from earlier rate equation treatments of the OGE[10,11] by explicitly calculating the electron temperature consistent with inelastic and superelastic collisions with excited states and by explicitly including different isotopes of the species of interest. The enrichment of copper ions (atomic masses 63 and 65) is investigated for conditions where the current is held constant and the $^2D_{5/2}$–$^2P_{3/2}$ transition at 510.6 nm in the 63-amu isotope is saturated by laser excitation. The degree of enrichment of the 63-amu ion is examined as a function of initial neutral abundance, discharge current density, discharge tube radius, and the rate of resonant charge exchange. We will find that ion enrichment factors as large as 10% can be obtained under typical discharge conditions. These results should be representative of the enrichment that one might obtain with OGE in other metal vapor-rare gas discharge.

II. Description of the Rate Equation Model

The choice of neon and copper for the discharge species in this study was predicated by the availability of the cross-section data necessary to compute the electron collision rates, by the generic nature of the Cu–Ne system as an ideal metal vapor-noble gas discharge, and by experience gained in modeling high repetition rate copper vapor lasers.[12] A full set of electron collision cross sections has been calculated for excitation between all the levels of copper considered here[13,14] as well as for ionization of the ground state.[15] For ionization of excited states the analytic forms of Deutsch are used.[16] The cross sections for the neon buffer gas are tabulated and well-known.[17] These cross sections are necessary to accurately model the distribution of copper atoms among the various higher-lying levels, which are the source for the stepwise optogalvanic enhanced ionization.

Six atomic levels and the ground-state ion of each copper isotope are included in the analysis as well as three atomic levels and the ground-state ion of the neon buffer gas. The neutral copper levels are the $^2S_{1/2}$ ground state, the $^2D_{5/2}$, $^2D_{3/2}$, $^2P_{1/2}$, and $^2P_{3/2}$ excited states, and a pseudostate located at 5.7 ev above the ground state (see Fig. 1). The pseudostate was chosen to represent higher-lying and displaced terms in the copper atom,[13] particularly important when stepwise ionization is a major source of ions.

In the rate equations below the following notation is used. The isotope k of species s (copper or neon) which is excited to state j has a density N_{sj}^k. The ion of isotope k of species s has a density N_{sI}^k. In the summations the lowest excited state is the ground state. The rate equation for N_{sj}^k is

$$\frac{\partial N_{sj}^k}{\partial t} = \sum_l N_{sl}^k r_{slj} n_e - \sum_l N_{sj}^k r_{sjl} n_e + \sum_n \sum_{l,m} N_{sm}^n N_{sl}^k r_{slmj} - \sum_n \sum_{l,m} N_{sl}^n N_{sj}^k r_{sjlm} - N_{sj}^k (N_m r_{PI} + N^+ r_{CE}) - \frac{D_s^k N_{sj}}{\Lambda^2} + n_e N_{sI}^k (r_{rr} + n_e r_{crr}) - \sum_l \left(N_{sj}^k - \frac{g_{sj}}{g_{sl}} N_{sl}^k \right) B_{sjl}^k I_{sjl}^k - \frac{N_{sj}^k}{\tau_{sj}} . \quad (1)$$

The first term of Eq. (1) accounts for selective electron impact excitation of N_{sj}^k from level N_{sl}^k with rate constant r_{slj}. The second term is for the depletion of N_{sj}^k by electron impact collisions. Electron impact excitation between all levels of a given isotope as well as electron impact ionization of all neutral levels is included in the analysis. The first two terms of Eq. (1) are the mechanism for the selective stepwise ionization of isotope k. The electron impact rate constants r_{sjl} were assumed to be the same for all isotopes. The third and fourth terms of Eq. (1) are for the atom–atom collisions, with rate r_{slmj} for a transition between levels l and m of species s, resulting in the excitation of N_{sj}^k. These collisions occur between both like and unlike isotopes. The result is a nonselective redistribution among all the isotopes of the laser energy initially deposited in a single isotope. The nonselective associative ionization reaction between excited states of neutral copper is included in this class of reactions. The fifth term of Eq. (1) is for the nonselective ionization of N_{sj}^k by Penning reactions (rate r_{PI}) with neon-excited states (N_m) and by charge exchange reactions (rate r_{CE}) with neon ions (N^+). The sixth term of Eq. (1) is for the loss of N_{sj}^k by diffusion with rate D_s^k and diffusion length Λ. For our purposes we can approximate $\Lambda = R/2.405$, where R is the radius of the discharge tube. The seventh term is for electron–ion recombination with rates r_{rr} (radiative recombination) and r_{crr} (collisional radiative recombination). Electron–ion recombination was assumed to populate only the highest-lying neutral state of the appropriate isotope. The eighth term of Eq. (1) is for transitions resulting from the absorption of laser radiation of intensity I_{sjl}^k by level N_{sj}^k changing to level N_{sl}^k (degeneracies g_{sj} and g_{sl}) with rate constant B_{sjl}^k. The last term of Eq. (1) is for spontaneous emission from N_{sj}^k with lifetime τ_{sj}. Radiation trapping factors are used where appropriate.

Equation (1) as written is applicable to the copper atoms. For the neon atoms, the fifth term for Penning and charge exchange reactions is explicitly included in the previous term, the seventh term for absorption of laser radiation is not used, and only one isotope is considered.

The conservation equation for the ion of isotope k of species s, N_{sI}^k, is

$$\frac{\partial N_{sI}^k}{\partial t} = \sum_l N_{sl}^k r_{slI} n_e + \sum_l (N_m r_{PI} + N^+ r_{CE}) N_{sl}^k + \sum_n \sum_{l,m} N_{sl}^k N_{sm}^n r_{slmI} - \sum_n \sum_l N_{sl}^n N_{sI}^k r_{sl}^{CE} + \sum_n \sum_l N_{sl}^n N_{sI}^k r_{sl}^{CE} - N_{sI}^k n_e (r_{rr} + r_{crr} n_e) - \frac{D_{sa}^k N_{sI}^k}{\Lambda^2}. \quad (2)$$

The first term of Eq. (2) is for the electron impact ionization of N_{sl}^k with rate r_{slI}. This is the only truly selective source of ionization for isotope k. All the other terms of Eq. (2) are for either nonselective ionization processes or recombination reactions. The second term of Eq. (2) is for nonselective Penning reactions and nonselective charge exchange collisions with the buffer gas. The third term of Eq. (2) is for the nonselective Penning reaction between excited states of like and unlike isotopes. The fourth and fifth terms are for the resonant and nonresonant charge exchange collisions between level l and the ion of like and unlike isotopes with rate r_{sl}^{CE}. These last two terms are most influential in setting an upper limit to the enrichment one might realize using the OGE method. Charge exchange reactions which mix the ions of different isotopes force the abundance of the ions toward their initial neutral values. The sixth and last terms of Eq. (2) are for the loss of ion N_{sI}^k by recombination and ambipolar diffusion (with rate constant D_{sa}^k), respectively. Diffusion is a selective loss mechanism since the diffusion constant is a function of the mass of the isotope.

From conservation of energy the rate equation for the Maxwellian electron temperature T_e is

$$\frac{\partial T_e}{\partial t} = \frac{2}{3}\frac{e^2E^2}{m_e \sum_l \nu_{cl}} - \sum_l \frac{(2m_e)}{M} \nu_{cl}(T_e - T_g) - \sum_s \sum_{m>l} \frac{2}{3} \varepsilon_{slm} N_{sl} r_{slm} + \sum_s \sum_{m<l} \frac{2}{3} \varepsilon_{slm} N_{sl} r_{slm}. \quad (3)$$

In Eq. (3) E is the longitudinal electric field, ν_{cl} is the electron momentum transfer collision frequency with species l, M_l is the mass of species l, m_e is the electron mass, T_g is the gas temperature, and ε_{slm} is the energy separation between levels l and m of species s. The terms of Eq. (3) are for energy gained from the applied electric field, thermalization to the gas temperature, inelastic energy losses in exciting N_{sl} to N_{sm}, and energy gained by superelastic relaxation of N_{sl} to N_{sm}.

The average gas temperature T_g was calculated by balancing the Joule heating of the gas by conduction to the wall,

$$T_g = T_w + \frac{jER}{2h} \quad (4)$$

where T_w is the wall temperature, j is the average current density, and h is the heat transfer coefficient (W/cm^2 K). When the current through the discharge is held constant, the longitudinal electric field in the plasma is a function of j and the impedence of the discharge. Writing the total current through the discharge as $I_o = \pi R^2 j$, one can show that[18]

$$E = \frac{I_o}{1.36\, n_e R^2 \mu_e e} \quad (5)$$

where μ_e is the electron mobility.

The figure of merit we will use for the degree of enrichment is the enrichment factor, β_i where

$$\beta_i = f_{Ii}/f_{Ni} - 1.0 \quad (6)$$

In Eq. (6), f_{Ii} is the fraction of ions in the discharge belonging to isotope i with respect to the other isotopes of the same species, and f_{Ni} is the analogous fraction for the initial neutral abundance.

We assume that the discharge is operating under constant current conditions obtained by connecting the discharge in a series with a high-impedance resistor and a dc power supply. The total current through the discharge is a function of the external circuit elements. Unless noted otherwise, the exciting radiation is specified to be sufficiently intense to saturate the transition of interest. The method of solution was to specify a current, assume a low ion density (10^9/cm^3), and integrate the rate equations described above until steady-state values were obtained. Since the solution converged to the steady state within a few tenths of a millisecond, this analysis is valid for pulsed excitation with pulse length less than a millisecond.

III. Results for the Enrichment of ^{63}Cu Ions

The choice of which isotope is selectively excited depends on the tunability of the exciting radiation, its bandwidth, the isotope shift of the species, and the broadening of each line. Due to the high tube temperature (T_w > 1450°C) required to obtain sufficient copper vapor, Doppler broadening causes the closer hyperfine components of the isotopes to overlap. For

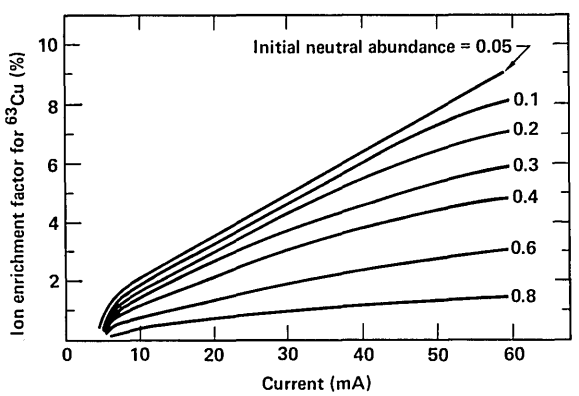

Fig. 2. Ion enrichment factor β for the 63-amu isotope of copper as a function of discharge current and initial neutral abundance of that isotope. The diameter of the discharge tube is 1 cm.

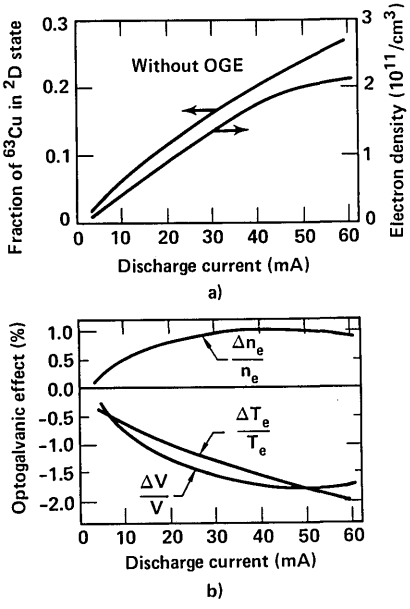

Fig. 3. Typical discharge parameters and OGE for the conditions of Fig. 2 and an initial 63-amu abundance of 0.3. The top figure shows conditions in the absence of the 510.6-nm radiation. The lower figure shows the change in electron density, electron temperature, and discharge voltage resulting from the presence of saturating 510.6-nm radiation. The electron temperature remains within a few percent of 1.2 eV for all current densities in the absence of the laser radiation.

the cases we will consider here, the most isolated transition, and hence the best choice for selective excitation, belongs to the 63-amu isotope of copper.[19] That is, we will attempt to enrich the fraction of 63-amu copper ions in the positive column discharge with respect to its neutral abundance by selectively exciting transitions in that isotope.

Selection of the lower-mass 63-amu component for enrichment is the worst case one can consider and, therefore, the most stringent test of using the OGE. Because of its lower mass, the ambipolar diffusion coefficient of the 63-amu ion is larger than the 65-amu ion. As a result, in the absence of optical excitation, the abundance of the 63-amu isotope is always slightly less than its neutral abundance (see below).[20] Therefore to achieve a net enrichment of the 63-amu isotope using the OGE method, this handicap must be overcome.

Although the natural abundance of ^{63}Cu is 0.69, in the discussion which follows the initial neutral abundance of ^{63}Cu is used as a free parameter. This allows us to address the general problem of isotope enrichment using the OGE over a large parameter space. The specific case of enrichment of natural copper is addressed by selecting the abundance 0.69.

A. β_{63} as a Function of Current Density

The calculated enrichment factor β_{63} as a function of discharge current and the initial abundance of the 63-amu isotope of copper is shown in Fig. 2. The exciting radiation is at 510.6 nm. The neon buffer gas pressure is 15 Torr, the total copper pressure is 0.1 Torr, and the wall temperature is 1600°C. The diameter of the discharge tube is 1 cm. For small initial abundances, β_{63} increases linearly with the discharge current density. Higher values of β_{63} are obtained with lower initial abundances. Although the electron temperature is low under our conditions ($\leq$1.5 eV), a significant fraction of the ions are produced by direct electron impact ionization of the ground-state or lower-lying levels of copper. The OGE is, therefore, competing with the ground state and ^2D ionization rate. The enrichment factor β_{63} is an indication of how successful this competition is. (Due to the disparity in ionization potentials, the fraction of ions belonging to the buffer gas is negligible.) When the selected isotope has a high neutral abundance, the fraction of ions which belong to that isotope and the ionization rate from the ground state are proportionally larger, and the OGE is less able to compete.

The increase in β_{63} as the current density increases has many contributing factors and one dominant cause. A contributing factor is due to a change in electron temperature. As the current density increases in the presence of the OGE radiation, the electron temperature decreases by as much as 5%. This decrease is a result of the decrease in the voltage drop across the discharge. (For typical discharge parameters and OGE signals, see Fig. 3.) The decrease in electron temperature decreases the nonselective rate of electron impact ionization from the ground state, thereby making stepwise ionization a larger contributor to the total ionization rate. This trend in electron temperature contrasts with the behavior observed in a hollow cathode discharge, where the electron temperature is less sensitive to the current density.

The dominant cause for the increase in β_{63} with increasing current density is an increase in the fraction of copper atoms in the $^2D_{5/2}$ state. As the current density increases, the fraction of copper atoms in the $^2D_{5/2}$ state increases (see Fig. 3). In the absence of 510.6-nm radiation, the fraction of atoms in the $^2P_{3/2}$ state is at least 2 orders of magnitude less than in the $^2D_{5/2}$ state. In the presence of intense 510.6-nm ra-

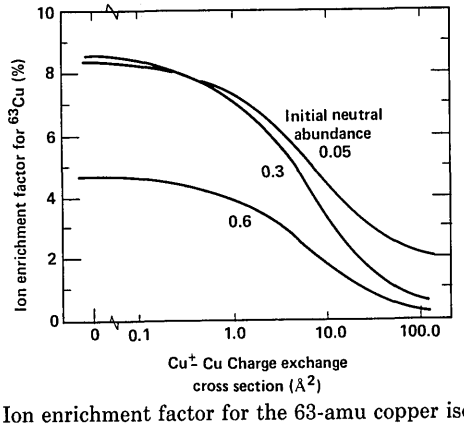

Fig. 4. Ion enrichment factor for the 63-amu copper isotope as a function of the value of the copper ion–copper charge exchange cross section and the initial neutral abundance of the 63-amu isotope. The current density was 25 mA/cm^2. The ion enrichment factors for low initial abundances are less sensitive to the deleterious effects of charge exchange.

diation, the density of atoms in the $^2D_{5/2}$ and $^2P_{3/2}$ states is driven to within a factor of 2 of being equal, thereby increasing the density of atoms in the $^2P_{3/2}$ state by more than an order of magnitude. Therefore, as the density of atoms in the $^2D_{5/2}$ state increases with increasing current, so does the density of atoms in the $^2P_{3/2}$ state. Consequently, the rate of stepwise ionization out of the $^2P_{3/2}$ state increases at nearly the same rate as the increase in the density of atoms in the $^2D_{5/2}$ state, thereby accounting for the increase in β_{63}.

B. Limiting Effects of Resonant Charge Exchange

The magnitude of the resonant charge exchange cross section between copper ions of different isotopes sets an upper limit on the enrichment factor β_{63}. In the limit where the rate of charge exchange is very large, the ionic abundances are clamped close to the value of the initial neutral abundances. The results of Fig. 2 were obtained using a resonant charge exchange cross section of 10 A^2. The sensitivity of β_{63} on this cross section for various initial abundances is shown in Fig. 4. The enrichment factor is not sensitive to interisotope charge exchange for cross sections of <1 A^2. A minimum value for the enrichment factor is obtained when the charge exchange cross section is >100 A^2. The enrichment factor for low initial abundances appears less sensitive to charge exchange than for higher initial abundances.

C. Geometry Effects

Due to the disparity in ambipolar diffusion rates between the two copper isotopes, the relative ion densities are a function of the radius of the discharge tube. This effect is illustrated in Fig. 5, where β_{63} is plotted as a function of the radius of the discharge tube while holding the current density constant. Examining the unperturbed discharge, the low-mass isotope, which has a larger diffusion loss rate, is depleted in the plasma. As the radius of the discharge tube increases, the relative diffusion loss rate decreases, and the disparity between ion and neutral abundances decreases.

In the presence of saturating radiation at 510.6 nm the enrichment factor β_{63} has a maximum value at a radius of ~0.6 cm. The decrease in β_{63} at smaller radii is a result of the inability of the OGE to compete with the large diffusion losses. The cause for the decrease in β_{63} at large radii is more subtle. As the radius of the discharge tube increases and the rate of ion diffusion decreases, the ion density increases. For otherwise constant conditions, however, the rate of mixing of the ions of the two isotopes by charge exchange collisions increases as the square of the ion density. As a result, over the range of radii considered in Fig. 5 the rate of decrease in β_{63} due to charge exchange increases by a factor of 15. It is this increase in charge exchange losses which accounts for the decrease in β_{63} at large radii.

D. Pressure Effects

In general, the enrichment factor β_{63} is independent of the partial pressure of copper when the neon pressure is held constant, provided the relative density of neon ions remains small. When the density of neon ions becomes a significant fraction of the total (greater than a few percent), β_{63} begins to decrease. This decrease in β_{63} is due in large part to a nonselective charge exchange of copper with neon ions. When holding the copper density fixed, higher pressures of neon in general increase β_{63}. This trend is a result of the higher neon pressure reducing the electron temperature, thereby making stepwise ionization a larger contributor to the total ionization rate compared with direct ground-state ionization.

E. Excitation by Resonance Radiation

For the conditions examined here the enrichment factor β_{63} increases as the intensity of 510.6-nm radiation increases and is maximum when the transition is saturated. When the saturating radiation is the 324.8-nm resonance line ($^2S_{1/2} - {}^2P_{3/2}$), there is often little, if any, enrichment. The enrichment factor β_{63} may in fact maximize with less than a saturating intensity. There are again many causes for this behavior. The OGE is significantly larger with the resonance radiation, and there is a corresponding larger increase in ion density. The large ion density significantly increases the copper–copper charge exchange rate, thereby canceling gains in selective ionization.

A major cause for the low ionic enrichment when exciting with resonance radiation is a consequence of the $^2D_{5/2}$ state being metastable. Under quasi-cw conditions the density of atoms in the $^2S_{1/2}$ and $^2P_{3/2}$ states has values proportional to the ratio of their degeneracies when saturated by resonance radiation. In the absence of other processes, one would expect a large

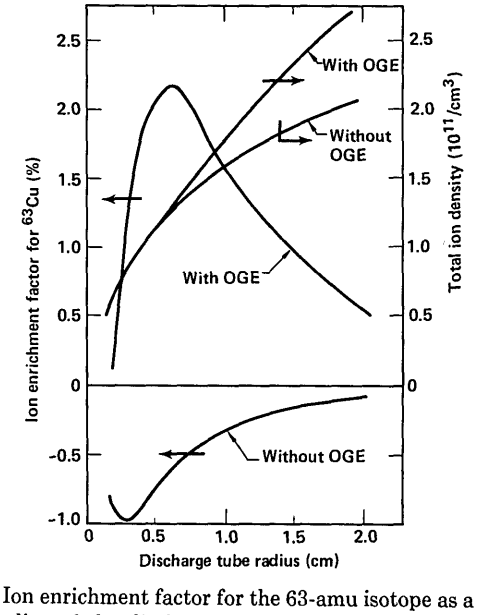

Fig. 5. Ion enrichment factor for the 63-amu isotope as a function of the radius of the discharge tube with and without saturating 510.6-nm radiation. The initial neutral abundance was 0.5, and the current density was held constant at 30 mA/cm^2. The extremum in β as the discharge tube radius increases results from competition between decreasing ion losses due to diffusion and increasing ion mixing from charge exchange collisions as the discharge tube radius increases. The charge exchange rate is proportional to the square of the ion density (also shown).

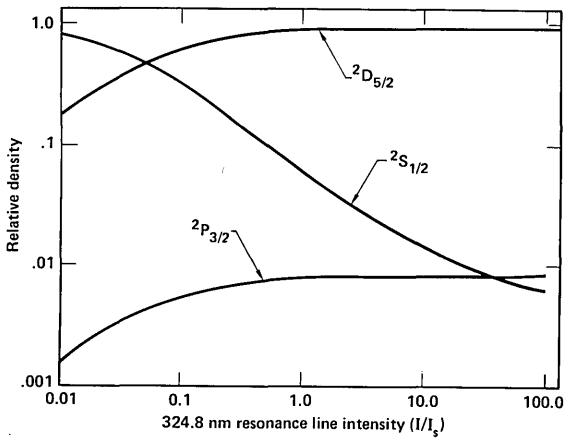

Fig. 6. Fraction of atoms of the 63-amu copper isotope in the $^2P_{1/2}$, $^2D_{5/2}$, and $^2P_{3/2}$ states as a function of the average intensity of radiation at 324.8 nm ($^2S_{1/2} - {}^2P_{3/2}$). (I_s is the saturation intensity.) The increasing fraction of atoms in the $^2D_{5/2}$ state reduces the contribution of stepwise ionization of this isotope relative to the unexcited isotope.

density ($\approx 10^{14}$ cm^{-3}) of atoms in the $^2P_{3/2}$ state. The $^2P_{3/2}$ state, however, is strongly coupled to the metastable $^2D_{5/2}$ state by both radiative decay and electron impact collisions. As a result, there is an efficient transfer of copper atoms from the ground state to the resonance level (via the OGE radiation) and then from the resonance level to the metastable level (via radiative decay and electron collisions). Atoms accumulate in the $^2D_{5/2}$ state, leading to a large density of atoms in that state ($\approx 10^{14}$/cm^3). The remaining smaller density of atoms ($\approx 10^{13}$/cm^3) is shared between the ground state and $^2P_{1/2}$ state (see Fig. 6). As a result, the stepwise ionization rate for that isotope relative to the unexcited isotope can actually decrease when excited by saturating resonance radiation, leading to small or negative ionic enrichment.

Although the details of the preceding discussion are particular to copper discharges, the general conclusions can be applied to any OGE system where the higher-lying state of the saturated transition has a large branching ratio to a lower metastable state. This situation effectively removes atoms from the rapid stepwise excitation process by storing atoms in a state with a relatively large ionization potential.

IV. Concluding Remarks

The isotopic enrichment of copper ions in the positive column of a Cu–Ne discharge has been examined with a rate equation analysis. The model, which can be considered generic for a metal vapor-noble gas discharge, predicts that isotopic enrichment of 10% is possible by using the optogalvanic effect. The parameter space in which this value can be achieved though is rather narrow. The isotope to be enriched must have a relatively low initial abundance (≤ 0.1) and the quasi-steady-state current density must be high (≥ 75 mA/cm^2).

The actual enrichment process requires an ion extraction technique which has not been addressed here but nevertheless sets an upper limit on the throughput of an enrichment system. Charge exchange reactions between ions of different isotopes as well as between the metal atoms and buffer gas ions set a fundamental as well as a systematic limit on the enrichment process. The fundamental limit is manifested by the fact that nonselective charge exchange collisions force the ion abundance toward the initial neutral values. The systematic limit is a result of the rate of charge exchange reactions increasing rapidly as the diameter of the discharge region increases, thereby limiting the volume scalability of the process.

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