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CHARGE-EXCHANGE EXCITATION AND cw OSCILLATION IN THE ZINC-ION LASER*

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We have investigated the afterglow decay rates of the Zn II laser lines in spontaneous emission. Measurements indicate that the dominant mechanism for excitation of these lines is thermal-energy charge-transfer collisions between zinc atoms and both atomic and molecular ions of helium.

We have previously proposed[1,2] that thermal-energy charge-transfer collisions involving the helium ion and ground-state zinc atoms of the type

$$He^+ + Zn \rightarrow He + (Zn^+)^* \qquad (a)$$

could be used to obtain cw oscillation in the zinc-ion laser and that such collisions in fact constituted the dominant mechanism for excitation of the upper levels in its earlier pulsed form.[3] Our subsequent investigations have given support to this proposal.

Relatively little work has been done at low energies involving inelastic processes such as Reaction (a). Lipeles, Novick, and Tolk[4] have reported charge transfer with simultaneous excitation in He^+-Ar reactions at beam energies as low as 10 eV with cross sections of about 10^{-16} cm^2. As discussed by Dworetsky et al.,[5] appreciable cross sections for this type of reaction may be expected since the levels of the temporary molecule formed during the collisions are grossly different from the levels of the isolated particles. Thus, for example, excitation can occur nonadiabatically through "curve crossings" between the different intermolecular potentials describing the molecule formed during the collision.

There is even less quantitative information available regarding inelastic charge-transfer reactions at thermal energies, and the experimental situation regarding optical excitation through thermal-energy charge-exchange reactions has not changed greatly since the qualitative review given sometime ago by Meyerott.[6]

During preliminary work with the zinc-ion laser, we observed the decay rates of the laser transitions in spontaneous emission in the afterglow of a pulsed discharge.[1] These decay rates were rather slow ($\simeq 10^4$ sec^{-1}) and increased linearly with zinc partial pressure. This was in sharp contrast to transitions whose upper states were slightly above the energy of the helium ion. The decay rates of the latter transitions were fast and did not change with zinc density. It was thus apparent that the upper laser states were being populated in the afterglow by inelastic collisions involving some long-lived carrier of excitation (see Fig. 1).

In a simplified model of the discharge afterglow, the carrier density will vary roughly as

$$N = N_0 \exp(-\gamma t), \qquad (1)$$

where

$$\gamma = D/\Lambda^2 + n\bar{v}\bar{\sigma}. \qquad (2)$$

The first term in γ represents diffusion to the walls with subsequent de-excitation; the second term represents a volume loss due to collisions with ground-state zinc atoms of density n. $\bar{v}$ is the mean relative thermal velocity between the carrier and the zinc atoms, $\bar{\sigma}$ is the total velocity-averaged cross section for destruction of the carrier, and Λ is the characteristic diffusion length of the container. To the extent that impurity ions excited by collisions such as those of Reaction (a) decay in a time short compared with $1/\gamma$, the light emitted by the impurity states would also be expected to vary as the carrier density. The above model assumes that only the dominant diffusion mode is excited, and that effects such as three-body collisions, collisions

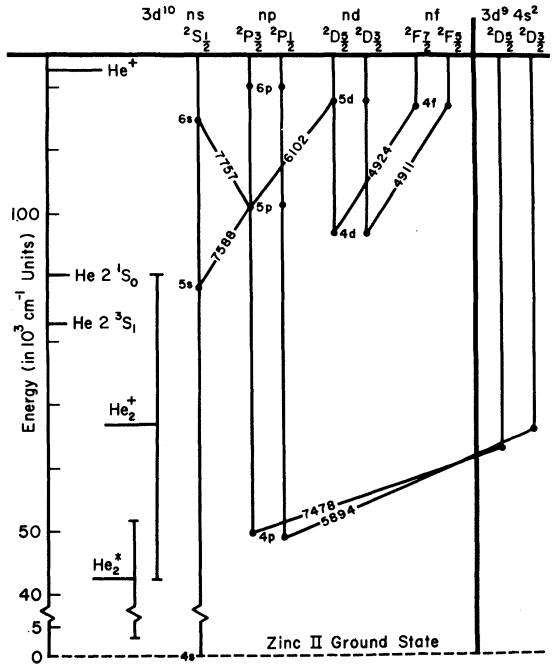

FIG. 1. Partial term diagram of Zn II indicating the laser transitions investigated in spontaneous emission. (The ionization limit of Zn II is not indicated.) The approximate energies available from the metastable particles in an inelastic collision are also shown. (The ground-state repulsive energy has been subtracted from the helium molecular states in the manner of Meyerott, Ref. 6.)

with other impurities, and significant depletion of the zinc ground-state atoms during the discharge do not occur. Care was taken in the present experiments to use sufficiently low discharge currents as to minimize the effects of such additional nonlinear processes.

The afterglow radiation was monitored with a photomultiplier tube, whose output was fed into a 100-channel analog wave-form averaging device with a maximum resolution of 1 μsec/channel. The output of the wave-form averager was converted into digital form and fed into a time-sharing computer which provided least-squares fits of the data by either a single- or a double-exponential functional form.

In order to avoid spurious effects from finite instrumental response time ($\simeq$2 μsec) and to insure adequate time for electron thermalization in the afterglow of the discharge (typically, $\lesssim$10 μsec), acquisition of the data was begun $\gtrsim$30 μsec after the current pulse. Under these conditions a single exponential decay gave a remarkably good fit to the data. Analysis of data taken earlier in the afterglow required two exponential components in order to achieve a sum of weighted squares for the fit compatible with the inherent noise level in the data. The slow component obtained in these two-component reductions always agreed with the single component obtained from analysis later in the afterglow. For all of the higher lying laser levels in Zn II, the fast time constant was consistently equal to the instrumental response time ($\simeq$2 μsec) and independent of the zinc density. However, for the $3d^9 4s^2$ terms the fast component ($\simeq$5 μsec) varied noticeably with the zinc density. This variation may have some physical significance in the formation processes of the energy carrier populating these states. However, it is impossible within the framework of the present experiment to isolate the nature of this process from other phenomena such as electron thermalization which would also be dependent on the zinc density and occur within the same time scale. Therefore, only the data for the slow component are reported in the present work and given physical significance in the interpretation.

We have made measurements of the decay rates of all of the Zn II laser transitions over a range of zinc densities from $\simeq 10^{12}$ to 10^{14} cm^{-3} and at helium pressures of 2, 4, and 8 Torr. The results at 4 Torr are shown in Fig. 2. Measurements were made by observing the emission from the end of a 1-cm-diam discharge tube 20 cm long. This tube was enclosed in an oven capable of maintaining a uniform temperature over the length of the discharge region. Zinc was distributed in small pockets along the bore of the tube and the zinc density was varied by controlling the temperature in the oven. The zinc partial pressure was determined from the discharge tube temperature and the relation[7]

$$\log_{10} P_{\mathrm{Torr}} = 8.741 - (6630^{\circ}\mathrm{K})/T. \qquad (3)$$

We have considered the possible involvement of five distinct metastable particles which exist in the afterglow of a pulsed helium discharge: the 2^1S_0 and 2^3S_1 atomic metastables; the He_2 $2s\,^3\Sigma_u^+$ state of the neutral helium molecule; the helium molecular ion, He_2^+; and the atomic helium ion, He^+. Figure 1 shows the approximate energies available from these particles in an inelastic collision[8,9] where the repulsive ground-state He_2 electronic energy has been subtracted off in the manner of Meyerott.[6] The latter would only be strictly valid in the limit that the Franck-Condon principle were obeyed in the inelastic collision

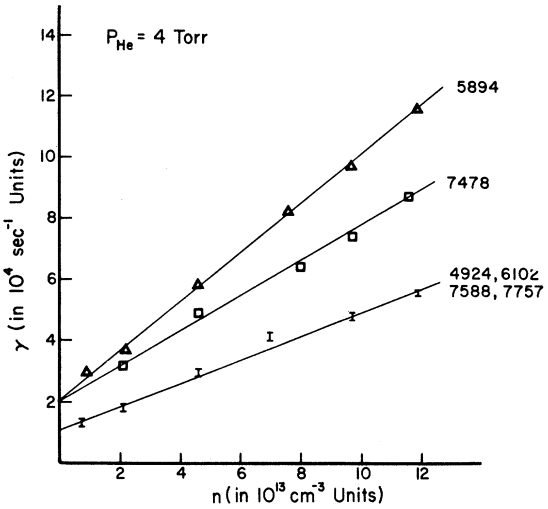

FIG. 2. Spontaneous-emission decay rates of the laser lines plotted against the zinc-atom density. The destruction cross sections determined from the 4924-, 6102-, 7588-, and 7757-Å transitions of Zn II were all approximately equal to 2.1×10^{-15} cm^2, assuming the relative velocity for the collision to be that for an atomic helium state. Assuming that a helium molecular state excited the Zn II states radiating on the 5894- and 7478-Å transitions, these data correspond to destructive cross sections of 6.1 and 4.4×10^{-15} cm^2, respectively.

between the excited helium molecule or molecular ion and the impurity. Because three heavy particles are involved in such a collision the approximation is very crude. Nevertheless, it seems probable that the Franck-Condon principle would be obeyed to some extent in grazing collisions of this type.

Excitation of the $4f\,^2F$, $5d\,^2D$, and $6s\,^2S$ states is the easiest to interpret. The atomic ion, He^+, is the only long-lived state which is energetically capable of exciting these levels. Also, the zero-zinc-density intercept of the γ plot for these transitions corresponds roughly to the calculated diffusion-loss rate of the helium ion.

The 7588-Å line from the $5p\,^2P_{3/2}$ state is fed by cascade transitions from the $5d\,^2D$ and $6s\,^2S$ states and it is therefore not surprising that this transition (7588 Å) shows a similar behavior to the 4924-, 6102-, and 7757-Å lines.

The data for the 5894- and 7478-Å transitions show a distinctly different behavior and imply that the $3d^9 4s^2$ ($^2D_{5/2}$, $^2D_{3/2}$) states are excited by a different carrier of energy. Ionizing collisions with metastable helium atoms (the Penning effect) has recently been suggested by Silfvast[10] as a dominant means for exciting the equivalent configuration ($4d^9 5s^2$) of Cd II in the helium-cadmium laser, and hence this process was considered here. Using absorption techniques, we have measured the decay rates of both of the atomic metastables as a function of zinc density. The 2^1S metastable decay rates were found to be very sensitive to slight traces of other impurities in the system whereas the decay rates of the Zn II lines were not. Although the 2^3S decay rates were comparable with those observed for the 4924-Å group, the 2^3S metastables do not have enough energy to excite the Zn II states involved. We therefore conclude that the Penning effect is not a dominant source of excitation in this system.

Another possible mechanism which was considered for the excitation of the $3d^9 4s^2$ configuration consisted of ionizing collisions involving neutral helium metastable molecules. To the extent that the Franck-Condon principle is satisfied in the collision process, the available energy from the metastable molecule is too low to excite these states unless it is in a high vibrational level. The metastable-neutral-molecule densities were much too low under our discharge conditions to permit a quantitative study of their relaxation rates in the afterglow of the discharge. In fact, the intensity of transitions terminating on these levels was strongly quenched by the presence of the zinc. These two observations imply that the metastable-molecule densities are not very significant in the helium-zinc discharges involved in the present work. Hence the neutral molecule does not appear to be an important source of Zn II excitation under these conditions. It is not an unlikely possibility that a primary source of metastable molecules occurs through recombination of He_2^+ molecular ions with electrons and that this source of excited neutral molecules is eliminated through charge-exchange collisions of the type

$$He_2^+ + Zn \rightarrow (Zn^+)^* + 2He \qquad (b)$$

in our experiment.

The He_2^+ molecular ion clearly has enough energy to excite the $^2D_{3/2}$ and $^2D_{5/2}$ levels and a literal interpretation of Meyerott's argument would predict that the He_2^+ ion was in very close resonance for Reaction (b). The fact that the decay rates for the 5894- and 7478-Å transitions have different slopes suggests that the $^2D_{3/2}$ and the $^2D_{5/2}$ states are excited by different carriers. A possible explanation for this difference is that both the first and second vibrational levels of the

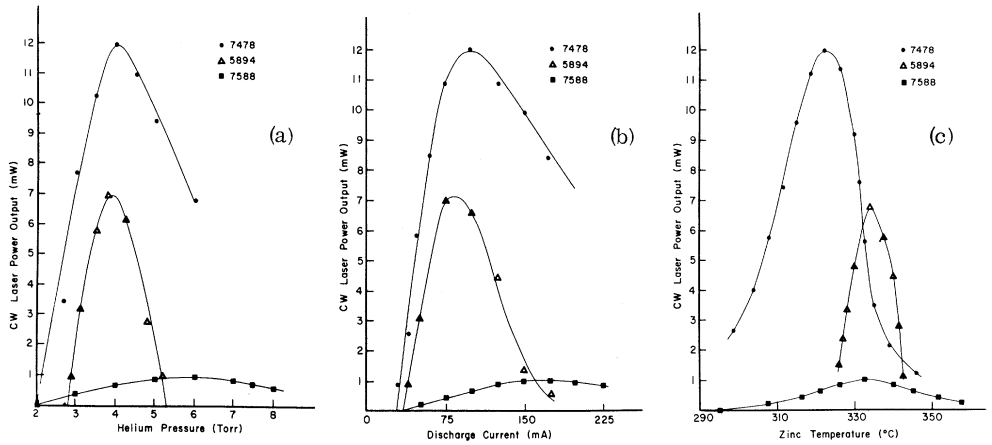

FIG. 3. Cw-laser output power as a function of (a) helium pressure, (b) discharge current, and (c) zinc temperature.

ground electronic state of He_2^+ are significantly populated and selectively transfer their excitation energy at different rates to the two $3d^9 4s^2$ states. The convergence of the two γ plots at zero zinc density is consistent with this assumption.

The group of $3d^9 4s^2 \rightarrow 3d^{10} 4p$ transitions is also of particular theoretical interest in that the upper state involves an inner-shell vacancy while the actual laser line occurs on an electric dipole transition that would be forbidden in the absence of configuration mixing. The $3d^9 4s^2$ configuration of the zinc ion would, of course, be excited with high probability in the "sudden" removal of a d electron from the $3d^{10} 4s^2$ ground state of neutral zinc, and the possible involvement of d-shell interactions in the excitation of mercury-, cadmium-, and zinc-ion lasers has been suggested before.[11,12] Consequently, a detailed investigation of these effects is presently being carried out.[13]

We have extended the work on the zinc-ion laser system by obtaining pulsed oscillation on two new laser transitions at 4911 and 5894 Å, as well as by achieving cw oscillation at 7478, 5894, and 7588 Å. The dominance of thermal-energy charge transfer in the zinc-ion laser provides a substantial improvement in the required current (typically 20 mA) for oscillation threshold as compared with noble-gas–ion lasers (typically a few amperes). Further, the use of ground-state ions as the initial carrier offers the possibility of substantial improvement in efficiency over the noble-gas–ion lasers.

The variation of laser output power on the three cw transitions with respect to helium pressure, zinc pressure, and discharge current is shown in Fig. 3. The cavity parameters have not been optimized. The mirror reflectance was about 99.8% at the laser wavelength and the active region of the discharge was 90 cm long and 4 mm in diameter. The power measurements made at optimum pressures under these conditions were 12, 7, and 1 mW for the 7478-, 5894-, and 7588-Å transitions, respectively.

Measured values of the lower state radiative lifetimes,[14] and our studies of the output spectrum, indicate that the natural width of the 5894-Å zinc-ion laser transition is comparable with the full Doppler width of the line. In contrast to all other known gas-laser transitions, it should be possible to use up most of the line in single-mode operation without relying on power or collision broadening. We are currently investigating this possibility.

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PLASMA DIFFUSION AND STABILITY IN TOROIDAL SYSTEMS*

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The problems of resistive diffusion and of rotation about the magnetic axis in toroidal systems are considered. They are shown to be related to the question of stability of drift-acoustic waves on which viscosity has an important influence. A nonrotating state is always unstable, but if the appropriate criterion is satisfied, there are stable rotating states for which an enhanced diffusion is calculated.

It is well known that the resistive diffusion rate for plasma in a toroidal system exceeds that in a similar straight system. The enhancement factor was first calculated by Pfirsch and Schluter[1] to be $1+8\pi^2/\iota^2$, where ι is the rotational transform, but recently[2] Stringer obtained a different result for a plasma rotating about its magnetic axis. As Stringer realized, his solution was not entirely satisfactory as the diffusion was not ambipolar and might be negative for some values of the rotational velocity $\vec{v}_0=(\vec{E}_0\times\vec{B})/B^2$.

We have re-examined the problem of diffusion in a toroidal system in the fluid model and shown that it is intimately related to the stability of collisional drift waves. In particular an equilibrium with zero rotation is unstable with respect to changes in the uniform rotation speed v_0. Furthermore, if resistivity were the only dissipative mechanism, as assumed by Stringer, then even if all deformations were artifically excluded there would be no stable value of v_0.

However, viscous stresses parallel to the magnetic field (hitherto ignored in both diffusion[2] and stability calculations[3]) alter this picture. When they are taken into account the state $v_0=0$ remains unstable, but if the stability criterion derived below is satisfied, there are two nonzero values of v_0 at which the equilibrium is stable both against changes in v_0 and against all axisymmetric[4] deformations. In each of these stable states, which are the only ones for which a calculation of diffusion is meaningful, the diffusion rate exceeds the Pfirsch-Schluter value by approximately

$$\left[1+1.2\left(\frac{m}{M}\right)^{1/2}\left(\frac{2\pi}{\epsilon\iota}\right)^2\left(\frac{r}{n}\frac{\partial n}{\partial r}\right)^2\right],$$

where $\epsilon=r/R$ is the aspect ratio of the torus.

The basic equations of our calculation are the full fluid equations with inclusion of resistivity η, viscosity ν, and the off-diagonal pressure tensor Π,[3,5] that is

$$-\nabla\varphi+\vec{v}\times\vec{B}=\eta\vec{j}-\frac{T_e}{ne}\nabla n+\frac{\vec{j}\times\vec{B}}{ne}, \tag{1}$$

$$nM[\partial\vec{v}/\partial t+\vec{v}\cdot\nabla\vec{v}] = -(T_e+T_i)\nabla n+\vec{j}\times\vec{B}+nM\nu\nabla^2\vec{v}-\nabla\cdot\Pi, \tag{2}$$

$$\partial n/\partial t+\nabla\cdot(n\vec{v})=0, \tag{3}$$

$$\nabla\cdot\vec{j}=0, \tag{4}$$

where φ is the potential, $\vec{j}$ the current, n the particle density, and $\vec{B}$ the magnetic field. The dominant effect of $-\nabla\cdot\Pi$ is equivalent to replacing $\vec{v}\cdot\nabla\vec{v}$ by $(\vec{v}-\vec{U}_i)\cdot\nabla\vec{v}$, where U_i is the ion diamagnetic velocity,[3,5] i.e.,

$$U_j\equiv\frac{T_j}{|e|B}\frac{1}{n}\frac{\partial n}{\partial r}.$$

The essential difference between these equations and those of Ref. 2 is in the inclusion of viscosity.

We apply these equations to an axisymmetric toroidal system of large aspect ratio, so that $\epsilon\ll1$, and work in the usual toroidal coordinate