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## Metallic ions in cometary comae and plasma tails

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Ion composition measurements made in the environment of the comet Giacobini-Zinner by the International Cometary Explorer (ICE) spacecraft have shown that  $\text{H}_2\text{O}^+$  is a major ion species, together with  $\text{CO}^+$  ions<sup>1</sup>. The presence of these two cometary ions has long been evident in optical spectra of cometary ion tails<sup>2</sup>. However, the ICE observations were surprising in that they detected ions of mass 23–24 AMU with a relatively high abundance<sup>1</sup>. According to the experimenters, these ions may be either  $\text{Na}^+$  or  $\text{C}_2^+$ , if not both. We suggest here that the ions detected may indeed be in part  $\text{Na}^+$  and/or  $\text{Mg}^+$  and that these and other metallic ions (especially  $\text{Si}^+$  and  $\text{Fe}^+$ ) may be an important component of the cometary ionosphere and central plasma tail. Our reasons are similar, in principle, to those which account for the prevalence of such ions in sporadic E layers in the terrestrial ionosphere, notably the comparatively short timescales for ionization of their neutral parent atoms and the large difference between the rates of dissociative and radiative recombination<sup>3,4</sup>.

Because of its strong optical emission, the source strength of  $\text{C}_2$  relative to that of  $\text{H}_2\text{O}$  and the scale length for photodissociation and/or photoionization have been well determined. The average values are:  $Q(\text{C}_2) \sim 4 \times 10^{-3} Q(\text{H}_2\text{O})$  and  $R(\text{C}_2) \sim 1.2 \times 10^5 \text{ km}$ , respectively<sup>5</sup>. In comparison, the corresponding scale length for the ionization of the water-group neutrals (that is,  $\text{H}_2\text{O}$ ,  $\text{OH}$  and  $\text{O}$ ) is a factor of 10 larger. It is possible, therefore, that the faster ionization of  $\text{C}_2$  radicals in a more limited region could lead to some enhancement of the relative abundance of  $\text{C}_2^+$  ions with respect to  $\text{H}_2\text{O}^+$  ions in a region  $< 10^5 \text{ km}$  from the tail axis.

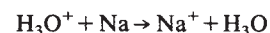
Sodium D-line emissions and emissions from other metallic species (Ca, Cr, Co, Mn, Fe, Ni, Cu, V, Si and K) are usually seen most prominently in Sun-grazing comets or bright comets at solar distances  $< 0.5 \text{ AU}$  (refs 6, 7). It is often argued that these emissions originate from atoms released by the sublimation of refractory dust grains<sup>8</sup>. At a solar distance of 1 AU, however, the sublimation rate of such dust grains is negligible and essentially no metal atoms should be released. Sodium D-line emissions were detected in comet West (1975n) when it was at a solar distance of 1.4 AU, which gave rise to the suggestion that a component of Na atoms could be embedded in volatile icy grains or matrix in atomic or molecular forms such as NaOH (ref. 9). Although estimates for the production rate of these Na

atoms (or their parent molecules) are very uncertain (an upper limit of  $Q(\text{Na})/Q(\text{H}_2\text{O}) \leq 10^{-3}$  may be set), such observations, together with the ICE ion composition measurement, call attention to the potential importance of  $\text{Na}^+$  ions and metallic ions, in general, in cometary comae and ionospheres. Below, we discuss several implications of metallic ion production in the coma environment and, in particular, emphasize that metallic ions could be a significant component of the central ionosphere and ion tail if their production rates are sufficiently large. The relative (solar) abundances of the most important species concerned are Na (0.06), Mg (1.00), Al (0.08), Si (0.95), Ca (0.07) and Fe (0.85)<sup>10</sup>.

Note that, in addition to sublimation from non-volatile dust grains and possibly from volatile material containing their parent molecules, sputtering of dust grains and the nucleus itself by solar-wind protons and the energetic 'pick-up' ion created in the coma should provide a source of secondary neutral atoms ( $\sim 90\%$ ) and ions ( $\sim 10\%$ )<sup>11</sup>. For a comet with a gas production rate up to that of Halley ( $Q = 10^{30}$  molecules  $\text{s}^{-1}$  at 1 AU) and a dust production rate  $\dot{m}_d/\dot{m}_g \sim 1$ , the effective cross-sectional area of the dust coma is estimated to be  $10^{17} \text{ cm}^2$  assuming a cutoff radius of  $1 \mu\text{m}$  (ref. 12). With a solar-wind flux of  $3 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$  and a sputtering yield of about unity, the effective sputtering rate of neutral atoms and ions from the dust coma is of the order of  $5 \times 10^{24} \text{ s}^{-1}$ , which is a factor of  $2 \times 10^5$  less than the volatile gas production rate. This alone might not be sufficient to account for the relatively high fluxes of ions of mass 23–24 detected by the ICE spacecraft at  $2 \times 10^4 \text{ km}$  from the tail axis. Nevertheless, secondary ions must be produced by sputtering in the coma and their fluxes may be detectable; there are processes which in any case tend to enhance their fluxes relative to those of molecular ions. Furthermore, the local production rates of sputtered atoms and ions may be larger than we have estimated here if  $m_d \gg m_g$  locally (as a consequence of differing expansion speeds of the gas and dust components) and if there is a significant component of dust particles with radii  $< 1 \mu\text{m}$ .

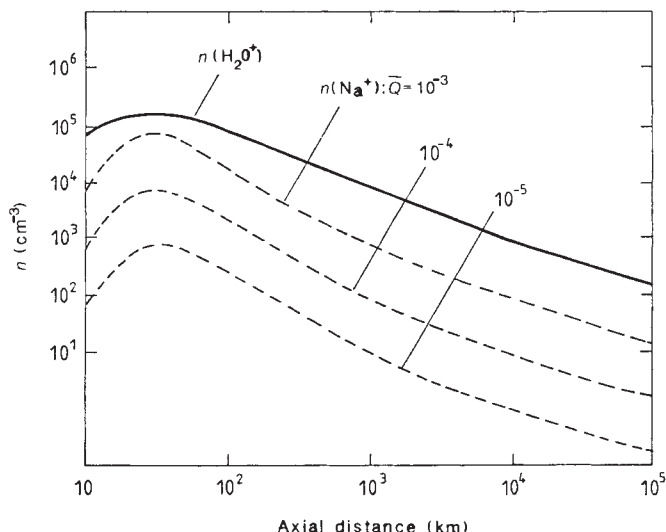
In general, the solar-wind plasma should not be able to reach the surface of the nucleus but the large gyroradii ( $\sim 1\text{--}5 \times 10^4 \text{ km}$ ) of energetic pick-up ions may allow them to do so. Thus the nucleus and inner coma should be continuously subject to sputtering even when the outgassing rate is large. However, because of the small area of the nucleus ( $\leq 10^{12} \text{ cm}^2$ ), the production rate of neutral atoms and ions arising from direct energetic particle sputtering is many orders of magnitude less than the sublimation rate of  $\text{H}_2\text{O}$  and other gases. Indeed, only when a comet nucleus is inactive at large solar distances would surface sputtering have a major role in releasing surface material into the surrounding coma region<sup>13,14</sup>.

Despite the relatively low production rates of sputtered atoms, it is important to recognize that by virtue of their short photoionization timescales ( $\sim 3 \times 10^4 \text{ s}$  at 1 AU (ref. 9) compared with  $3 \times 10^6 \text{ s}$  for  $\text{H}_2\text{O}$ ), together with the fact that a substantial fraction of the sputtered gas is already ionized, metallic ions could become relatively enriched. The very fact that such neutral atomic species as Na appear in the spectra of some comets suggests that their photoions should be abundant. The ionization potentials of these atoms are such that they are photoionized by hydrogen Lyman- $\alpha$  radiation, whereas the more abundant molecules require photons of 12 eV or more for photoionization. In addition to photoionization, metallic ions can be produced by charge transfer reactions such as



which has a rate coefficient  $\leq 10^{-9} \text{ cm}^3 \text{ s}^{-1}$  (ref. 15); this process may be important in the denser regions of the cometary ionosphere where the  $\text{H}_3\text{O}^+$  density is  $\sim 10^4 \text{ cm}^{-3}$ .

It is difficult to find reactions which can neutralize metallic ions faster than radiative recombination, which with a typical recombination coefficient of  $10^{-12} \text{ cm}^3 \text{ s}^{-1}$  is itself unimportant in the present context. To remove metallic ions quickly they



**Fig. 1** Comparison of the number density of the  $\text{H}_2\text{O}^+/\text{H}_3\text{O}^+$  ions with that of  $\text{Na}^+$  (or other metallic ions) for different relative source abundances of the metallic neutrals,  $\bar{Q} = Q(\text{Na})/Q(\text{H}_2\text{O})$ . In the calculation, simulating the free-streaming ionospheric flow along the tail, the flow speed increases linearly from  $1 \text{ km s}^{-1}$  near the nucleus to  $100 \text{ km s}^{-1}$  at  $3 \times 10^4 \text{ km}$  from the nucleus. The metal ion enhancement in this case is entirely the result of the relatively high ionization rate of their atoms in comparison with that of water molecules.

must first be converted to some form of molecular ion so that dissociative recombination is a possibility. However, the rates of formation of such molecular ions are low, except perhaps very close to the nucleus: for example, in the case of  $\text{Na}^+$  the process



has a rate coefficient of only  $1.6\text{--}6.1 \times 10^{-16} \text{ cm}^3 \text{ s}^{-1}$  at 20 K (ref. 16). This is competitive with radiative recombination in regions where the  $\text{H}_2\text{O}$  density is  $10^4$  or more (that is, within  $3 \times 10^4 \text{ km}$ ) but provides loss rates for  $\text{Na}^+$  ions which are significantly only in regions where the  $\text{H}_2\text{O}$  density exceeds  $10^9 \text{ cm}^{-3}$  and the electron density is  $\geq 10^2 \text{ cm}^{-3}$ .

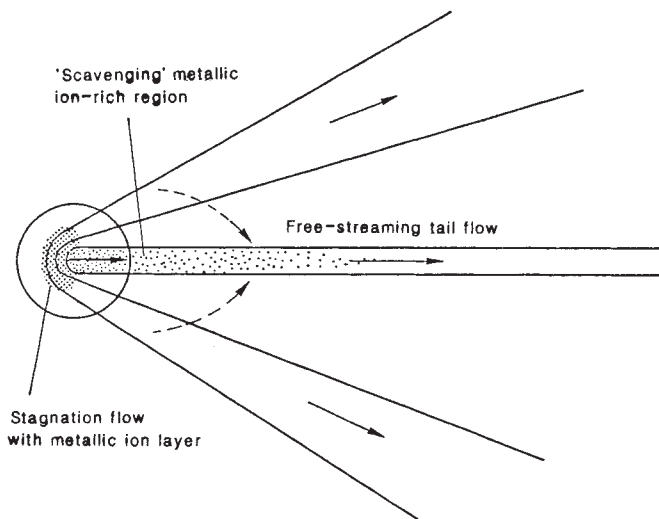
In the inner coma, a basic (convective) timescale is determined by the plasma flow characteristics. These are, in turn, controlled by drag which resists motions of plasma relative to the neutrals and by the tension stress created by strongly curved magnetic field lines which pull the plasma inwards towards the nucleus on the upstream side and eventually out along the tail on the downstream side<sup>17</sup>. The flow speeds in the 'ionospheric' region surrounding the nucleus may be quite small, perhaps  $10^2\text{--}10^3 \text{ m s}^{-1}$ , corresponding to convective timescales of  $10^4\text{--}10^5 \text{ s}$ . The tail plasma flow behind the nucleus can be regarded to a first approximation as an expansion of the ionospheric plasma along a quasi one-dimensional tunnel, accelerated by the thermal pressure gradient and magnetic field streams into a region of negligibly small neutral drag. In this manner, flow speeds of  $10\text{--}30 \text{ km s}^{-1}$  may be achieved in  $\sim 3 \times 10^4 \text{ km}$ , giving a convective timescale of  $\sim 10^3\text{--}10^4 \text{ s}$ . Detailed calculations are required to analyse these processes fully, involving the ion chemistry as well as the plasma flow dynamics, but the essential features can be demonstrated by a simple approach.

Figure 1 shows results from a model calculation with a range of the relative production rate of the metallic atoms ( $Q(\text{Na})/Q(\text{H}_2\text{O}) = 10^{-5}, 10^{-4}$  and  $10^{-3}$ ). Ionospheric plasma flow in the tailward direction is simulated here by converting to a one-dimensional geometry with the flow acceleration approximated by assuming  $d(\log A)/dx = (2/x) \exp(-x/X_a)$  for the stream-tube area  $A$  and an acceleration  $dV/dx = (100 \text{ km s}^{-1})/X_b$ . The scale lengths are taken to be  $X_a = 3 \times$

$10^3 \text{ km}$  and  $X_b = 3 \times 10^4 \text{ km}$ . This calculation demonstrates that an enhancement of a factor of  $\sim 100$  could be introduced into the metal ion abundance simply as a result of more rapid ionization.

There is a second important effect which could enrich the metallic ion component of the cometary plasma: in the sunward region of the ionosphere, which is a region of very low speed flow, metallic ions carried in or produced *in situ* could gradually replace  $\text{H}_2\text{O}^+$  as the dominant ion as a consequence of their differing rates of recombination (see Fig. 2). The lifetime of ions against recombination is  $\tau \sim 1/\alpha N_e$ , where  $\alpha$  is the recombination coefficient and  $N_e$  the electron density. For metallic and other atomic ions the (radiative) recombination coefficients are typically of the order of  $10^{-12} \text{ cm}^3 \text{ s}^{-1}$ , whereas for molecular ions, which undergo dissociative recombination,  $\alpha \sim 10^{-6} \text{ cm}^3 \text{ s}^{-1}$ . According to magnetohydrodynamic (MHD) simulations (J. A. Fedder, personal communication), ions produced in or brought into the central coma reside for as long as  $10^5 \text{ s}$  with electron densities of  $10^2\text{--}10^4$  or more; in these circumstances the metallic ions simply accumulate ( $\tau \sim 10^8\text{--}10^{10} \text{ s}$ ), whereas molecular ions continually recombine ( $\tau \leq 10^2\text{--}10^4 \text{ s}$ ). Thus, as plasma is carried through the coma by contraction of the looped magnetic field lines to eventually form the plasma tail of the comet, it should become progressively enriched in atomic, especially metallic, ions to a much greater extent than would be implied by their higher ionization rates alone. The formation of such a metallic ion-rich zone by 'magnetic scavenging' in the neutral atmosphere of a comet is analogous to the occurrence of sporadic E in the Earth's ionosphere, where, following rapid photoionization of metallic atoms, neutral atmospheric wind shears in the presence of the geomagnetic field produce the scavenging effect. This, in turn, leads to the formation of metallic ion-rich layers of high plasma density in which the molecular ions may actually be depleted relative to the surroundings<sup>3,4,18,19</sup>.

The ICE spacecraft passed through the tail of comet Giacobini-Zinner along a path very favourable for the detection of metallic ions but unfortunately it was not designed for this purpose and so only a hint of their presence may have been obtained. However, the Giotto spacecraft has five mass spectrometers of various types which may be able to detect and identify metallic ions. Such ions should be most prevalent in the region immediately surrounding the nucleus ( $< 10^4 \text{ km}$ ) but



**Fig. 2** A schematic view of how the metallic ions might be accumulated in the low-speed flow region of the cometary ionosphere. Metallic ions produced in the coma upstream from the nucleus are carried inwards and concentrated, without suffering recombination losses as do molecular ions, before eventually escaping downstream along the tail.



some should be detectable at larger distances. In some cases, the metallic ions may be submerged beneath large peaks associated with more abundant ions but  $\text{Fe}^+$ , in particular, may be relatively free of such interference and hence be more easily identifiable.

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## Gas-like nature of the sodium anion in solution

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During the past decade a considerable amount of evidence has accumulated for the existence of an anion of sodium,  $\text{Na}^-$ , as a stable, long-lived entity in certain non-aqueous solvents<sup>1-7</sup>. The term sodium anion refers to a spherically symmetric diamagnetic species in which, in the orbital model of atomic structure, the two valence electrons are accommodated in the 3s orbital of sodium<sup>7</sup>. We report here direct measurements of  $^{23}\text{Na}$  nuclear spin-lattice relaxation rates for  $\text{Na}^-$ , coupled with nuclear magnetic resonance (NMR) chemical shift data, which are used to show that  $\text{Na}^-$  in solution is almost completely decoupled from its environment, behaving as if it were in the gas phase.

The systems chosen for study were solutions of sodium and mixed sodium/alkali metal samples ( $\text{Na}/\text{M}$ , where  $\text{M} = \text{K}, \text{Rb}$  and  $\text{Cs}$ ) in the solvent 1, 4, 7, 10-tetraoxacyclododecane (12-crown-4, 12C4; Fig. 1). This solvent has a powerful and specific ability to complex alkali cations. For example, the dissociation of sodium/potassium alloy proceeds through the disproportionation reaction<sup>8</sup>



where  $\text{K}_s^+$  represents a complexed or solvated potassium cation.

Metal solution samples were prepared under stringent high-vacuum conditions<sup>8</sup> and  $^{23}\text{Na}$ ,  $^1\text{H}$  and  $^{13}\text{C}$  relaxation rates were measured using established procedures.

The frequency of the  $^{23}\text{Na}^-$  resonance was found to be unchanged to within 0.5 p.p.m. on examining, over a range of temperatures, about 50 metal/12C4 solutions differing in both concentration and counter cation. All these  $\text{Na}^-$  resonance frequencies correspond to an increase in the electronic shielding

of the  $^{23}\text{Na}$  nucleus relative to that in the gaseous sodium atom,  $\text{Na}_{\text{gas}}^0$ , by  $\sim 2.5$  p.p.m., after allowing for bulk-susceptibility corrections. To within the experimental errors arising from relating the solution measurements to the  $\text{Na}_{\text{gas}}^0$  data, the shift in resonance frequencies is the same as that predicted (2.88 p.p.m.) from an accurate calculation<sup>7</sup> of the shielding of a gaseous sodium anion,  $\text{Na}_{\text{gas}}^-$ , relative to that of  $\text{Na}_{\text{gas}}^0$ . This indicates that both the 2p and 3s electrons of  $\text{Na}^-$  in solution interact very weakly, if at all, with their surroundings, the resonance frequency of the  $\text{Na}^-$  ion being diamagnetically shifted relative to that of  $\text{Na}_{\text{gas}}^0$  by the additional shielding generated by the two, spin-paired 3s valence electrons.

The shielding of  $\text{Na}^-$  relative to  $\text{Na}_{\text{gas}}^0$  should be contrasted with the substantial de-shielding of  $\sim 60$  p.p.m. of  $\text{Na}_s^+$  relative to  $\text{Na}_{\text{gas}}^0$ , which is considerably larger than the 5.18 p.p.m. de-shielding<sup>7</sup> of a gaseous sodium cation,  $\text{Na}_{\text{gas}}^+$ , relative to  $\text{Na}_{\text{gas}}^0$ . The additional de-shielding of  $\text{Na}^+$  in the solution arises from the Ramsey paramagnetic contribution to the shielding generated by the interaction of  $\text{Na}^+$  with the environment. Any such paramagnetic contribution is absent in the gaseous atoms and ions.

A detailed examination<sup>9</sup> of the concentration and temperature dependence of the rates of  $^{23}\text{Na}$  nuclear spin-lattice relaxation shows that the dominant, but still extremely inefficient, relaxation arises from the interaction of the nuclear electric quadrupole moment ( $eQ$ ) of the  $^{23}\text{Na}$  nucleus with fluctuating electric field gradients. The relaxation of nuclei with spin ( $I$ )  $> 1/2$  is expected to be dominated by this quadrupolar mechanism<sup>10</sup>.

Nuclear spin-lattice relaxation rates ( $T_{1n}^{-1}$ ) for  $\text{Na}^-$  in 12C4 are very much less than the value of  $3,770 \text{ s}^{-1}$  for  $\text{Na}^+$  in this solvent. The cation relaxation arises from the very large fluctuations in the electric field gradient caused by the asymmetric environment of the  $\text{Na}^+$  ion resulting from its strong complexation by the solvent<sup>11</sup>. The very small relaxation rate for  $\text{Na}^-$  in the solvent 12C4 is comparable with that reported in Table 1 for  $\text{Na}^+$  in  $\text{H}_2\text{O}$ , which experiences an environment, the time average of which is centrosymmetric<sup>10,12,13</sup>.

It is therefore of interest to compare the  $\text{Na}^-$  nuclear relaxation rate with those of other ions which are in a centrosymmetric environment<sup>10,12,13</sup>. Note, however, that the experimental spin-lattice relaxation times of Table 1 are not, at this stage, directly

Table 1 Measured and scaled nuclear spin relaxation rates for ions in various solvents

Ion/solvent	(1)	(2)	(3)	(4)
	Measured $T_{1n}^{-1}$ * ( $\text{s}^{-1}$ )	Nuclear properties ( $I, eQ$ )	$T_{1n}^{-1}$ scaled for: Electronic anti-shielding ( $\beta_2$ )	Viscosity ( $\tau_{\text{sol}}$ )
$\text{Na}^+/\text{H}_2\text{O}$	16.2	16.2	191	7,109
$\text{Na}^-/12\text{C4}$	3.33	3.33	3.33	3.33
$\text{Cl}^-/\text{H}_2\text{O}$	25	39	1.66	61.66
$\text{Cl}^-/\text{C}_2\text{H}_5\text{OH}$	1,300	2,047	86	716.7
$\text{Cl}^-/12\text{C4}$	14,100	22,260	935	935
$\text{I}^-/\text{H}_2\text{O}$	4,600	346	3.58	133
$\text{Cs}^+/\text{H}_2\text{O}$	0.075	817	15.5	577

\* With the exception of relaxation rates in 12C4 these data are taken from refs 12, 13.

comparable because  $T_{1n}^{-1}$  is proportional, among other things, to the square of the nuclear electric quadrupole moment ( $eQ$ )<sup>2</sup>, and depends on a theoretically well-established function of the nuclear spin  $I$ . Thus, column (2) of Table 1 reports the suitably scaled  $T_{1n}^{-1}$  values that would arise if all the ions had the same  $eQ$  and  $I$  as the  $^{23}\text{Na}$  nucleus. This scaling (from column (1) to (2) in Table 1 uses theory of such firmly established validity<sup>10</sup> that these numbers can be regarded as experimental results.

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