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CYANIDE DEPOSITS IN CO_2 TEA LASERS USING AMINE ADDITIVES

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A B S T R A C T

Samples of the deposits formed around the discharge region in CO_2 TEA lasers operated with amine additives have been analysed by infra-red spectrophotometry. The deposits contain appreciable amounts of cyanides, and liberate hydrogen cyanide when treated with acid.

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A considerable number of authors (see reference list) have described the beneficial effects of various amine additives in CO_2 TEA lasers. The use of tri-methylamine as an additive in excimer lasers has also recently been reported (Bezójian, 1976). These additives have low ionization potentials, and provide improved pre-ionization when used in conjunction with UV spark sources (Seguin, 1976 and 1977 for example).

Lasers operated in our laboratories with tri-ethylamine and tri-methylamine additives have produced considerable quantities of a yellow buff deposit which accumulates around the discharge region, but especially near the sliding-arc UV pre-ionization source. This source consists of an array of brass washers fixed with epoxy resin to a glass substrate, with a copper earth plane similarly fixed on the other side of the glass.

A sample of the deposit was incorporated in a KBr disc and analysed by infra-red spectrophotometry, and the results compared with reference spectra. The analysis indicated appreciable amounts of cyanide and/or cyanate groupings, the best matches being with metal cyanides, but did not rule out the presence of organic nitriles and isonitriles. Infra-red analysis of the gases evolved when a portion of the deposit was treated with hydrochloric acid showed HCN to be present, which tends to confirm the presence of a metal cyanide in the deposit. The absorption spectra of the solid also indicated the presence of carbonates, nitrate, silica and possibly amine or amide species.

The deposit arises mainly from decomposition of the amine additives, the metal cyanides being formed by reaction with metal vapour from the sliding-arc electrodes, which are slowly eroded. The expoxy resin is not thought be to involved in the process as the deposit occurs in other TEA lasers in which no expoxy is used.

Although the analysis was not quantitative, appreciable amounts of cyanide compounds are present. Appropriate caution should therefore be exercised when dismantling or cleaning TEA lasers in which these additives have been used. Acid cleaning results in the liberation of HCN, and should be avoided.

The laser from which these deposits were taken had been operated primarily with tri-ethylamine and to a lesser extent with tri-methylamine, but it seems likely that cyanide-containing deposits will form in lasers using any of the amine additives.

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THE HELICAL FLUX AND OTHER INTEGRAL INVARIANTS IN A SLIGHTLY DISSIPATIVE PLASMA

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THE HELICAL FLUX AND OTHER INTEGRAL INVARIANTS IN A SLIGHTLY DISSIPATIVE PLASMA

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In the present short note the Hamiltonian approach followed in a preceding paper [1] for the derivation of the approximate conservation of the helical flux in a toroidal slightly dissipative plasma is extended and applied to all particles and not only to those on a resonant surface. The procedure leads to a helical invariant and to other integral invariants which hold under more general conditions than the adiabatic invariant considered in Ref. [1].

We consider a toroidal plasma subject to a small dissipation resulting from slight collisional effects on an originally unperturbed axisymmetric equilibrium formed by magnetic surfaces with circular cross sections. The motion of the particles can be described in a canonical formalism with the following Hamiltonian:

$$H = \Sigma_j \frac{1}{2M_j} \left(\vec{p}_j - \frac{e_j}{c} \vec{A}(\vec{x}_j,t) \right)^2 + \lambda \Sigma_{ji} \frac{e_j e_i}{|\vec{x}_j - \vec{x}_i|} \tag{1}$$

Here the index j labels the quantities related to the j-th particle; λ is a smallness parameter indicating that the Coulomb interaction is considered as a small perturbation. Besides an external magnetic field, the potential $\vec{A}(\vec{x},t)$ also includes the magnetic field created by the particles through the current $\vec{j}(\vec{x},t) = \Sigma_j (e_j/c) \dot{\vec{x}}_j(t)\, \delta(\vec{x} - \vec{x}_j(t))$ and the Maxwell equations. In general in the unperturbed equilibrium a macroscopic current exists, resulting from an organized drift motion of the particles with uniform velocity, whose collective effect is included in $\vec{A}$ at the order $\lambda = 0$. The vector potential $A(\vec{x}_j,t)$ experienced by the j-th particle is sensitive to the other particles also through the effect of the Coulomb interaction, which affects the velocities and the trajectories of the particles only at the order λ .

We introduce a curvilinear coordinate system $x^i \equiv (r,\theta,\varphi)$ in which the unperturbed lines of force are the straight lines $\theta = \varphi/q(r) + \text{const}$ ($q(r)$ is the safety factor). It is however convenient to pass from the

θ,φ variables to new W_o, w_o variables through the canonical transformation

$$P = n p_2 + m p_3 \quad , \quad W_o = \frac{\theta}{n}$$
$$p = mq\, p_3 \; , \; w_o = \frac{n\varphi - m\theta}{qmn} \qquad (2)$$

where m,n are integers and the p_i are the covariant components of the canonical momenta $p_i = M\dot{x}_i + e\, A_i(x^i, t)/c$ in the $x^i \equiv (r,\theta,\varphi)$ coordinate system. The canonical equations of motion for the j-th particle then take the form:

$$\dot{P}_j = -\frac{\partial H}{\partial W_{oj}} \quad , \quad \dot{W}_{oj} = \frac{\partial H}{\partial P_j}$$
$$\dot{p}_j = -\frac{\partial H}{\partial w_{oj}} \quad , \quad \dot{w}_{oj} = \frac{\partial H}{\partial p_j} \qquad (3)$$

We now introduce the quantity

$$\overline{P}_j \equiv \int_0^{2\pi} P(r_j, W_{oj}, w_{oj}, t)\, dW_{oj} \qquad (4)$$

and observe that, apart from an additive contribution related to the inertial term $M\dot{x}_i$ of the canonical momenta and considering r and w_o as constants (the index j will be dropped for simplicity wherever this can be done without confusion), $\overline{P}$ is the magnetic flux $-\chi$ (neglecting henceforth the factor e/c) across a helical ribbon enclosed by the axis $(r = 0)$ and by the helix $r = \text{const}$, $n\varphi - m\theta = \text{const}$ (or $w_o = \text{const}$) , covered from $\theta = 0$ to $\theta = 2\pi n$:

$$-\chi(r, w_o, t) = \frac{1}{n} \int_0^{2\pi n} (n A_2 + m A_3)\, d\theta \qquad (5)$$

This interpretation is however only valid when r and w_o are constant. At the contrary, in the definition (4) of $\overline{P}$ the (r, W_o, w_o) are time-dependent lagrangian variables; hence, in order that $\overline{P}$ (after neglecting the inertial terms) has the meaning of a helical flux, as defined above, one must have sensitively $r(t) \approx \text{const}$, $w_o(t) \approx \text{const}$ during the time needed by a particle to cover the range of definition of $\overline{P}$, namely the interval between $W_o = 0$ (or $\theta = 0$) and $W_o = 2\pi$ (or $\theta = 2\pi n$). This implies certain restrictions on the particles and the process to be considered. Indeed noting that in the small Larmor radius approximation the unperturbed motion of the circulating particles occurs along a line of force, one obtains at zero order:

$$\dot{w}_o = \frac{nq - m}{nm\, q^2} \dot{\varphi} \tag{6}$$

It follows that $w_o(t) \approx \text{const}$ only in the neighbourhood of a resonant surface $q \approx m/n$. Moreover one has $r(t) \approx \text{const}$ only when the characteristic time τ in which $r(t)$ changes sensitively satisfies the condition

$$\tau \gg \frac{2\pi n r}{v_\theta} = \frac{2\pi n R q}{v_{th}} \tag{7}$$

where R is the major radius of the torus and $\mathbf{v}_\theta$ is the poloidal velocity of a particle moving along a line of force; v_θ is related to the thermal velocity by the relation $v_\theta \approx v_{th}\, r/qR$. As was shown in Ref. [1], under the preceding limitations, $\overline{P}$ can be identified with the adiabatic invariant J, apart from terms of second order in the Coulomb interaction.

It is now our purpose to discuss the conservation of $\overline{P}$ without using the two restrictions indicated above. Indeed in this case it will be shown that $\overline{P}$, although it cannot be interpreted as a magnetic flux, is nevertheless still conserved under slightly more restrictive conditions for the interaction. Let us proceed to calculate the time derivative of $\overline{P}$ using the canonical equations (3):

$$\begin{aligned} \dot{\overline{P}} &= \int_0^{2\pi} \left(\frac{\partial P}{\partial w_o} \dot{w}_o + \frac{\partial P}{\partial r} \dot{r} + \frac{\partial P}{\partial t} \right) dW_o = \int_0^{2\pi} \left(\dot{P} - \frac{\partial P}{\partial W_o} \dot{W}_o \right) dW_o \\ &= - \int_0^{2\pi} \left(\frac{\partial H}{\partial W_o} + \frac{\partial P}{\partial W_o} \frac{\partial H}{\partial P} \right) dW_o \end{aligned} \tag{8}$$

Noting that

$$\frac{dH}{dW_o} = \frac{\partial H}{\partial W_o} + \frac{\partial H}{\partial P}\frac{\partial P}{\partial W_o} + \frac{\partial H}{\partial p}\frac{\partial p}{\partial W_o} \tag{9}$$

the expression for $\dot{P}$ takes the form

$$\dot{P} = -\int_0^{2\pi} \left(\frac{dH}{dW_o} - \frac{\partial H}{\partial p}\frac{\partial p}{\partial W_o} \right) dW_o = \int_0^{2\pi} \dot{w}_o \frac{\partial p}{\partial W_o} dW_o \tag{10}$$

where use has been made of the fact that H is a periodic function of W_o. In order to discuss the order of magnitude of this expression for $\dot{P}$ we consider a situation in which the relevant effect of the collisions consists in driving a relatively short time scale magnetic process (as for instance tearing modes, disruptive processes, field reversal in pinches) the time scale being much shorter than that of the normal electric dissipation. We assume that the overall effect of this process on the magnetic part of the Hamiltonian consists in the superposition of a time dependent helical perturbation (i.e. depending on t and w_o) on the original toroidal axi-symmetric equilibrium. The Hamiltonian for the particle motions of the system can then be written in the form

$$H = \Sigma_j [H_o(P_j, p_j, r_j, W_{oj}) + \eta H_1(P_j, p_j, r_j, W_{oj}, w_{oj}, t)] + \lambda \sum_{ij} \frac{e_j e_i}{|\vec{x}_j - \vec{x}_i|} \tag{11}$$

where η represents the amplitude of the collisionally driven magnetic perturbation. The dependence of H_o and H_1 on W_o, only results, in the limit $\lambda = 0$, from the toroidicity of the geometry and is then of order $\epsilon = r/R$. In general this dependence is $O(\epsilon) + O(\lambda)$. Now in order to assess the magnitude of $\dot{P}$, we take into account the following facts : i) At zero order in η and λ (unperturbed axisymmetric equilibrium) $p \approx mq(r) A_3(r)$ is independent from W_o. Indeed $-2\pi A_3(r)$ represents the poloidal flux, which only depends on r. So the lowest order dependence of p on W_o

occurs at the orders η and λ . ii) p is a periodic function of W_o and at the order $\lambda = 0$ one has $\partial p/\partial W_o = O(\epsilon\eta)$. iii) Finally we observe that the integral (10) can be different from zero only when the explicit dependence on W_o of $\dot{w}_o = \partial H/\partial p$ is taken into account. At the lowest order this dependence is $O(\epsilon) + O(\lambda)$. It follows from these facts that

$$\dot{\overline{P}} = O(\epsilon^2\eta) + O(\epsilon^2\eta^2) + O(\lambda\epsilon) + O(\lambda\eta\epsilon) + O(\lambda^2) \qquad (12)$$

So, if we neglect the individual effects of the collisions $(\lambda \to 0)$ and only take into account their overall effect on the magnetic behaviour of the system, we obtain that $\overline{P}$ is conserved apart from terms of order $\epsilon^2\eta$ and is then exactly conserved in the cylindrical limit $(\epsilon \to 0)$. If we only consider the particles on a resonant magnetic surface $(q = m/n)$, $\dot{w}_o$ vanishes at zero order in η and λ (see Eq. (6)) and $\overline{P}$ is now conserved apart from terms of order $\epsilon^2\eta^2$. This result agrees with that of Ref. [1], where $\overline{P}$ was treated as an adiabatic invariant.

On the ground of the foregoing result and of the canonical formalism one can readily establish the existence of integral invariants of the kind proposed by J.B. Taylor [2]. First we observe that $\overline{P}$ represents an area in the P,W_o space so that one has

$$\int_0^{2\pi} P dW_o = \int_{\overline{P}} dP dW_o = - \chi(r,w_o,t) \qquad (13)$$

More generally we shall start from a finite volume element in phase space

$$I \equiv \int_{\Delta w_o(t)} dw_o \int_{\Delta p(t)} dp \int_{\overline{P}} dP\, dW_o = -\int_{\Delta w_o(t)} dw_o \int_{\Delta p(t)} dp\, \chi(r,w_o,t) \qquad (14)$$

which is a constant of motion, according to the Liouville theorem, provided Δw_o and Δp depend on time according to the equations of motion. In order to calculate the value of I we take in Eq. (14) the initial instant of time and suppose that at this time the helical perturbation is so small that one can neglect the w_o dependence. In this case it is convenient to express I in terms of the toroidal flux Φ and the poloidal flux Ψ , which, in the case of an axisymmetric system, are approximated as follows:

$$\begin{aligned} \Phi &\equiv \int_0^{2\pi} A_2\, d\theta = 2\pi (A_2)_{\epsilon=0} + O(\epsilon^2) \\ \Psi &\equiv - \int_0^{2\pi} A_3\, d\varphi = -2\pi A_3 \end{aligned} \tag{15}$$

From the canonical transformation (2) (in which $q(r)$ is treated as a parameter) one obtains the relation $dp = mqd\,p_3 \approx mqd\,A_3 = -mqd\Psi/2\pi$, so that the invariant I takes the form

$$I = \frac{m\Delta w_{oo}}{2\pi} \int_{\Delta\Psi_o} q\,\chi\, d\Psi \tag{16}$$

where Δw_{oo} and $\Delta\Psi_o$ are the initial values of $\Delta w_o(t)$ and $\Delta\Psi(t)$. We now express the integral in terms of the variable Φ , remembering that one as $\Psi = \Psi(\Phi)$ with $qd\Psi = d\Phi$. Extending the integration over the whole plasma volume, Eq. (16) takes the form

$$I = \frac{m\Delta w_{oo}}{2\pi} \int_{\Phi_{oS}} \chi d\Phi \tag{17}$$

where Φ_{oS} is the initial value of Φ on the plasma surface S . The meaning of the preceding integral is clear when one remembers that, in the axisymmetric limit, χ is expressed by the relation

$$-\chi(\Phi) = n\Phi - m\Psi(\Phi) \tag{18}$$

Other integral invariants can be easily derived from Eqs. (17) and (18). For instance we consider the quantity

$$K_\Psi \equiv \int_{\Phi_{oS}} \Psi d\Phi = \frac{n}{2m}\Phi_{oS}^2 + \frac{2\pi}{m^2\Delta w_{oo}} I \tag{19}$$

Let us assume that Φ_{oS} is invariant, as is the case when the surface S is a metallic wall. In this case Φ_{oS} is independent from w_o at all times and from the invariance of the phase volume $\int_{\Delta w_o(t)} d w_o \int_{\Phi_{oS}} d\Phi$ one obtains that Δw_o is also invariant and equal to its initial value Δw_{oo}. The invariance of K_Ψ then follows from the fact that the r.h.s. of Eq. (19) is all expressed in terms of invariants.

A further invariant can be defined by means of a partial integration,

$$K_\Phi \equiv \int_{\Psi_{oS}} \Phi d\Psi = \Phi_{oS} \Psi_{oS} - K_\Psi \tag{20}$$

provided both Ψ_{oS} and Φ_{oS} are assumed invariant on the metallic surface S .

Finally, subtracting K_Ψ from K_Φ and using Eq. (15) (neglecting terms of order ϵ^2) one obtains the Taylor invariant:

$$K = - K_\Psi + K_\Phi = (2\pi)^2 \int (A_3 \, d A_2 - A_2 \, d A_3) = \int \vec{A} \vec{B} \, dV \tag{21}$$

In conclusion, rapid time dependent processes driven by a small amount of collisions in a large aspect ratio ($\epsilon \to 0$) toroidal plasma are describable up to first order in the Coulomb interaction in terms of the same constants of motion as for a particle subject formally to a helical time dependent electromagnetic field. Applying the Hamiltonian formalism one can derive, under certain restrictions, an invariant $\overline{P}$ which, in combination with the Liouville theorem, gives rise to the integral invariants considered above.

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