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Electron Transport and Rate Coefficients in Townsend Discharges

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Abstract

Electron swarm parameters such as the drift velocity, diffusion coefficients and ionization rates have been calculated by many authors. However, the values obtained for most parameters depend upon the type of experiment under consideration, so that, for a given gas and a fixed value of E/N, differences are obtained for steady-state Townsend, pulsed Townsend and time-of-flight experiments. It is shown that this is an unnecessary complication since each experiment can be analysed in terms of time-of-flight parameters. It is proposed that calculated parameters should be presented in a manner which would assist not only in the analysis of the above experiments but also in other swarm studies in progress.

1. Introduction

Thomas (1969) suggested that the value of a swarm parameter is different in steady-state Townsend discharges from the value applicable to a pulsed Townsend experiment. This idea has been developed in a systematic manner by Tagashira *et al.* (1977) for steady-state Townsend (SST), pulsed Townsend (PT) and time-of-flight (TOF) experiments. Subsequently, several authors have published calculated values of SST, PT and TOF parameters while in other instances values are quoted without reference to any particular experimental system.

The introduction of these various coefficients has highlighted the need for care when comparing calculated and observed quantities. However, many of these parameters are unnecessary since

(*a*) both SST and PT experiments can be analysed in terms of TOF parameters;

(*b*) published values of experimentally observable transport coefficients and ionization rates are all TOF parameters (other quantities such as the first Townsend ionization coefficient α_T can be expressed in terms of TOF parameters);

(*c*) the SST and PT parameters are related to TOF parameters.

In the present paper the relationship between TOF and other transport parameters is discussed. To facilitate comparison with the work of Tagashira *et al.* (1977) a one-dimensional swarm under uniform electric field conditions is analysed using a two-term Legendre polynomial expansion for the electron velocity distribution function.

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2. The Continuity Equation

The macroscopic electron swarm parameters are connected to the collision cross sections by means of the Boltzmann equation in which the electron distribution in configuration and velocity space is described by the probability distribution $f(\boldsymbol{r}, \boldsymbol{v}, t)$. In the two-term Legendre series expansion this is represented as

$$f(\boldsymbol{r}, \boldsymbol{v}, t) = f_0(\boldsymbol{r}, v, t) + f_1(\boldsymbol{r}, v, t)\,\mathrm{P}_1(\cos\theta),$$

where $\theta = 0$ is in the direction of the local convective velocity of electrons with speed v. The electron concentration is then

$$n(\boldsymbol{r}, t) = \int_0^\infty 4\pi v^2 f_0(\boldsymbol{r}, v, t)\,\mathrm{d}v\,.$$

By following standard procedures, an equation for $f_0(\boldsymbol{r}, v, t)$ can be obtained [c.f. Huxley and Crompton (1974), equation (5.41)]:

$$\frac{\partial f_0}{\partial t} - \nabla\boldsymbol{.}\left(\frac{v^2}{3\nu_{\mathrm{m}}}\nabla f_0 + \frac{ve\boldsymbol{E}}{3m\nu_{\mathrm{m}}}\frac{\partial f_0}{\partial v}\right) + \frac{1}{4\pi v^2}\frac{\partial}{\partial v}\{\sigma_{\mathrm{E}}(v) - \sigma_{\mathrm{coll}}(v)\}$$
$$= \{\nu_{\mathrm{i}}(v) - \nu_{\mathrm{a}}(v)\}f_0\,, \qquad (1)$$

where $\nu_{\mathrm{m}} = NQ_{\mathrm{T}}\,v$, $\nu_{\mathrm{i}}(v) = NQ_{\mathrm{i}}\,v$ and $\nu_{\mathrm{a}}(v) = NQ_{\mathrm{a}}\,v$ (assuming a two-body attachment process). Here N is the neutral gas concentration, Q_{i} and Q_{a} are the ionization and attachment cross sections, and Q_{T} is the sum of the elastic momentum transfer cross section and all inelastic cross sections (implying isotropic scattering for inelastic collisions). The last term on the left-hand side of this equation represents the rate-of-change of number density in an element $\mathrm{d}\boldsymbol{v}$ of velocity space. The details of this term are not of importance in this work since it does not contribute to the continuity equation in configuration space. Electron creation and loss processes are explicitly contained in the right-hand side of the equation.

Multiplying equation (1) by $4\pi v^2\,\mathrm{d}v$ and integrating over all speeds, we obtain

$$\partial n(\boldsymbol{r}, t)/\partial t + \nabla\boldsymbol{.}\boldsymbol{\Gamma}(\boldsymbol{r}, t) = n(\boldsymbol{r}, t)\{\nu_{\mathrm{i}}(\boldsymbol{r}, t) - \nu_{\mathrm{a}}(\boldsymbol{r}, t)\}\,,$$

where the particle flux is

$$\boldsymbol{\Gamma}(\boldsymbol{r}, t) = -\tfrac{4}{3}\pi\boldsymbol{E}\,\frac{e}{m}\int_0^\infty \frac{v^3}{\nu_{\mathrm{m}}}\frac{\partial f_0}{\partial v}\,\mathrm{d}v - \nabla\left(\tfrac{4}{3}\pi\int_0^\infty \frac{v^4}{\nu_{\mathrm{m}}} f_0\,\mathrm{d}v\right)$$

and

$$\nu_{\mathrm{i,a}}(\boldsymbol{r}, t) = 4\pi\int_0^\infty v^2\,\nu_{\mathrm{i,a}}(v) f_0\,\mathrm{d}v/n(\boldsymbol{r}, t)$$

are the local ionization and attachment rates.

The particle flux can be written as

$$\boldsymbol{\Gamma}(\boldsymbol{r}, t) = n(\boldsymbol{r}, t)\,\boldsymbol{W}(\boldsymbol{r}, t) - \nabla\{n(\boldsymbol{r}, t)\,D(\boldsymbol{r}, t)\}\,,$$

where the 'local' drift velocity and diffusion coefficient are defined by

$$W(\boldsymbol{r},t) = -\tfrac{4}{3}\pi\frac{e}{m}\boldsymbol{E}\int_0^\infty \frac{v^3}{v_\mathrm{m}}\frac{\partial f_0}{\partial v}\,\mathrm{d}v/n(\boldsymbol{r},t),$$

$$D(\boldsymbol{r},t) = \tfrac{4}{3}\pi\int_0^\infty \frac{v^4}{v_\mathrm{m}}f_0\,\mathrm{d}v/n(\boldsymbol{r},t).$$

For a one-dimensional swarm with the electric field $\boldsymbol{E}$ in the $-z$ direction, the continuity equation becomes

$$\frac{\partial n(z,t)}{\partial t}+\frac{\partial}{\partial z}\{n(z,t)\,W(z,t)\}-\frac{\partial^2}{\partial z^2}\{n(z,t)\,D(z,t)\} = n(z,t)\{v_\mathrm{i}(z,t)-v_\mathrm{a}(z,t)\}. \quad (2)$$

It remains to determine $f_0(z,v,t)$ from equation (1). This can be accomplished in the hydrodynamic regime by expanding the distribution in a series of powers of the spatial gradient of the number density (Skullerud 1969). In the following work it is assumed that $f_0(z,v,t)$ is known and, after transforming to the kinetic energy $\frac{1}{2}mv^2 = \varepsilon$ as an independent variable, can be written as:

$$f_0(z,\varepsilon,t) = n(z,t)\,G_0(\varepsilon)-\frac{1}{N}\frac{\partial n(z,t)}{\partial z}G_1(\varepsilon)+\frac{1}{N^2}\frac{\partial^2 n(z,t)}{\partial z^2}G_2(\varepsilon)-\dots, \quad (3)$$

with

$$\int_0^\infty G_0(\varepsilon)\,\mathrm{d}\varepsilon = 1 \qquad \text{and} \qquad \int_0^\infty G_n(\varepsilon)\,\mathrm{d}\varepsilon = 0, \qquad n\neq 0;$$

and $G_n(\varepsilon)$ are known functions. The factors $1/N$, $1/N^2$, ... are explicitly included to make the $G_n(\varepsilon)$ independent of gas concentration (Parker and Lowke 1969). If we put $V_1 = (2/m)^{\frac{1}{2}}$, then

$$n(z,t)\,W(z,t) = -\tfrac{1}{3}V_1\frac{Ee}{N}\int_0^\infty \frac{\varepsilon}{Q_\mathrm{T}}\frac{\partial}{\partial\varepsilon}\{\varepsilon^{-\frac{1}{2}}f_0(z,\varepsilon,t)\}\,\mathrm{d}\varepsilon, \quad (4a)$$

$$n(z,t)\,D(z,t) = \frac{V_1}{3N}\int_0^\infty \frac{\varepsilon^{\frac{1}{2}}}{Q_\mathrm{T}}f_0(z,\varepsilon,t)\,\mathrm{d}\varepsilon, \quad (4b)$$

$$n(z,t)\,v_{\mathrm{i,a}}(z,t) = V_1\,N\int_0^\infty Q_{\mathrm{i,a}}\,\varepsilon^{\frac{1}{2}}f_0(z,\varepsilon,t)\,\mathrm{d}\varepsilon. \quad (4c)$$

Using equations (3) and (4), equation (2) can be expressed in the following manner:

$$\frac{\partial n}{\partial t}+\frac{\partial}{\partial z}\Big(nW_0-D_\mu\frac{\partial n}{\partial z}+S_\mu\frac{\partial^2 n}{\partial z^2}-\dots\Big)-\frac{\partial^2}{\partial z^2}\Big(nD_0-S_\mathrm{D}\frac{\partial n}{\partial z}+\dots\Big)$$

$$= n(v_{\mathrm{i}0}-v_{\mathrm{a}0})-(W_\mathrm{i}-W_\mathrm{a})\frac{\partial n}{\partial z}+(D_\mathrm{i}-D_\mathrm{a})\frac{\partial^2 n}{\partial z^2}-(S_\mathrm{i}-S_\mathrm{a})\frac{\partial^3 n}{\partial z^3}+\dots, \quad (5)$$

where the constant coefficients are defined by

$$\nu_{\mathrm{i0,a0}} = V_1 N \int_0^\infty Q_{\mathrm{i,a}}\, \varepsilon^{\frac{1}{2}} G_0(\varepsilon)\, \mathrm{d}\varepsilon\,, \qquad \text{(dimension } \mathrm{T}^{-1}\text{)}$$

$$W_0 = -\tfrac{1}{3} V_1 \frac{Ee}{N} \int_0^\infty \frac{\varepsilon}{Q_\mathrm{T}} \frac{\partial}{\partial \varepsilon} \{\varepsilon^{-\frac{1}{2}} G_0(\varepsilon)\}\, \mathrm{d}\varepsilon\,,$$

$$W_{\mathrm{i,a}} = V_1 \int_0^\infty Q_{\mathrm{i,a}}\, \varepsilon^{\frac{1}{2}} G_1(\varepsilon)\, \mathrm{d}\varepsilon\,, \qquad \text{(dimension } \mathrm{L\,T}^{-1}\text{)}$$

$$D_0 = \frac{V_1}{3N} \int_0^\infty \frac{\varepsilon^{\frac{1}{2}}}{Q_\mathrm{T}} G_0(\varepsilon)\, \mathrm{d}\varepsilon\,,$$

$$D_\mu = -\frac{V_1}{3N} \frac{Ee}{N} \int_0^\infty \frac{\varepsilon}{Q_\mathrm{T}} \frac{\partial}{\partial \varepsilon} \{\varepsilon^{-\frac{1}{2}} G_1(\varepsilon)\}\, \mathrm{d}\varepsilon^{\frac{1}{2}}, \qquad \text{(dimension } \mathrm{L}^2\, \mathrm{T}^{-1}\text{)}$$

$$D_{\mathrm{i,a}} = \frac{V_1}{N} \int_0^\infty Q_{\mathrm{i,a}}\, \varepsilon^{\frac{1}{2}} G_2(\varepsilon)\, \mathrm{d}\varepsilon\,,$$

$$S_\mathrm{D} = \frac{V_1}{3N^2} \int_0^\infty \frac{\varepsilon^{\frac{1}{2}}}{Q_\mathrm{T}} G_1(\varepsilon)\, \mathrm{d}\varepsilon\,,$$

$$S_\mu = -\frac{V_1}{3N^2} \frac{Ee}{N} \int_0^\infty \frac{\varepsilon}{Q_\mathrm{T}} \frac{\partial}{\partial \varepsilon} \{\varepsilon^{-\frac{1}{2}} G_2(\varepsilon)\}\, \mathrm{d}\varepsilon\,, \qquad \text{(dimension } \mathrm{L}^3\, \mathrm{T}^{-1}\text{)}$$

$$S_{\mathrm{i,a}} = \frac{V_1}{N^2} \int_0^\infty Q_{\mathrm{i,a}}\, \varepsilon^{\frac{1}{2}} G_3(\varepsilon)\, \mathrm{d}\varepsilon\,.$$

3. Application to Experiment

The continuity equation as expressed by equation (5) can be applied to the analysis of various situations.

(*a*) *TOF Experiment*

In this experiment $n(z,t)$ or some closely related quantity is measured. The appropriate differential equation for $n(z,t)$ is obtained from equation (5) by collecting together terms in $n(z,t)$ and its derivatives to obtain

$$\frac{\partial n}{\partial t} + W \frac{\partial n}{\partial z} - D_\mathrm{L} \frac{\partial^2 n}{\partial z^2} + S \frac{\partial^3 n}{\partial z^3} + \text{(higher order derivatives)} = n(\nu_{\mathrm{i0}} - \nu_{\mathrm{a0}})\,, \tag{6}$$

where the TOF transport parameters are

$$W = W_0 + W_\mathrm{i} - W_\mathrm{a}\,,$$

$$D_\mathrm{L} = D_0 + D_\mu + D_\mathrm{i} - D_\mathrm{a}\,,$$

$$S = S_\mathrm{D} + S_\mu + S_\mathrm{i} - S_\mathrm{a}\,.$$

These parameters correspond to the TOF transport parameters given by Tagashira *et al.* (1977) [cf. equation (11) of their paper with $A_n = 0$ and $F_n(\varepsilon) = (-1/N)^n G_n(\varepsilon)$], in addition to the rate coefficients ν_{i0} and ν_{a0}. For an isolated swarm, W_0 is the instantaneously averaged velocity of all the electrons in the swarm, while W is the velocity of the centre-of-mass of the swarm. This W differs from W_0 since ionization adds relatively more electrons at the front of the swarm (cf. Blevin *et al.* 1978), while attachment may either increase or decrease the centre-of-mass velocity relative to W_0 depending on the energy dependence of the attachment cross section (if three-body attachment occurs then W will show a pressure dependence also). The 'isotropic diffusion coefficient' D_0 is equal to the transverse diffusion coefficient D_T in a three-dimensional swarm in the absence of ionization or attachment. However, production or loss processes will modify D_T so that it is no longer equal to D_0 (Tagashira *et al.* 1977). The difference between the longitudinal diffusion coefficient D_L and D_0 is produced by spatial variations in the electron mobility D_μ and ionization and attachment rates D_i and D_a. The rate coefficients ν_{i0} and ν_{a0} are swarm-averaged quantities.

Usually TOF experiments are analysed by neglecting terms of higher order than the second spatial derivative of n in equation (6), i.e.

$$\frac{\partial n}{\partial t} + W\frac{\partial n}{\partial z} - D_L\frac{\partial^2 n}{\partial z^2} = n(\nu_{i0}-\nu_{a0}), \tag{7}$$

with a solution for an isolated source at $z = 0$ and $t = t_0$,

$$n(z,t-t_0) = n_0 \exp\{(\nu_{i0}-\nu_{a0})(t-t_0)\}$$
$$\times \exp[-\{z-W(t-t_0)\}^2/4D_L(t-t_0)]/\{4\pi D_L(t-t_0)\}^{\frac{1}{2}}.$$

To the same degree of approximation, an SST experiment can be modelled by integrating this distribution over all source times $-\infty < t_0 \leqslant t$ (Huxley and Crompton 1974), giving in the one-dimensional case

$$n(z) = c\exp(\alpha_T z), \tag{8}$$

where c is a constant.

The value of α_T obtained by this procedure can be obtained by using equation (8) in (7) to give

$$\alpha_T = \frac{W}{2D_L} - \left\{\left(\frac{W}{2D_L}\right)^2 - \frac{\nu_{i0}-\nu_{a0}}{D_L}\right\}^{\frac{1}{2}}, \tag{9}$$

so that the SST parameter α_T can be expressed in terms of TOF parameters.

(*b*) *SST Experiment*

An alternative approach to the SST experiment can be given by using equation (8) in (5) to give

$$n(z,t)\,W(z,t) = n(z,t)(W_0 - \alpha_T D_\mu + \alpha_T^2 S_\mu - \ldots)$$
$$= n(z,t)\,W_{SST},$$

where

$$W_{\rm SST} = W_0 - \alpha_{\rm T} D_\mu + \alpha_{\rm T}^2 S_\mu - \dots . \tag{10}$$

Similarly, we get

$$D_{\rm SST} = D_0 - \alpha_{\rm T} S_{\rm D} + \dots, \tag{11}$$

$$(\nu_{\rm i})_{\rm SST} = \nu_{\rm i0} - \alpha_{\rm T} W_{\rm i} + \alpha_{\rm T}^2 D_{\rm i} - \alpha_{\rm T}^3 S_{\rm i} + \dots, \tag{12a}$$

$$(\nu_{\rm a})_{\rm SST} = \nu_{\rm a0} - \alpha_{\rm T} W_{\rm a} + \alpha_{\rm T}^2 D_{\rm a} - \alpha_{\rm T}^3 S_{\rm a} + \dots . \tag{12b}$$

The continuity equation then becomes

$$W_{\rm SST}\frac{{\rm d}n}{{\rm d}z} - D_{\rm SST}\frac{{\rm d}^2 n}{{\rm d}z^2} = n(\nu_{\rm i} - \nu_{\rm a})_{\rm SST}.$$

Again, using equation (8) we have

$$\alpha_{\rm T} = \left(\frac{W}{2D}\right)_{\rm SST} - \left\{\left(\frac{W}{2D}\right)^2_{\rm SST} - \left(\frac{\nu_{\rm i} - \nu_{\rm a}}{D}\right)_{\rm SST}\right\}^{\frac{1}{2}}. \tag{13}$$

This apparently gives a different result from equation (9). However, $\alpha_{\rm T}$ is contained in the definitions of the SST parameters and if terms involving S and higher derivatives are neglected (as assumed in the TOF case), the values of $\alpha_{\rm T}$ obtained from (9) and (13) are identical. Since $W_{\rm SST}$, $D_{\rm SST}$ and $(\nu_{\rm i,a})_{\rm SST}$ are not experimentally observable quantities it would seem that their continued use is not warranted.

(c) PT Experiment

This experiment depends on the motion of an electron swarm between two electrodes and the manner in which this changes the external circuit parameters. If the inter-electrode capacitance is C and the external series resistance is R, the experiment can be carried out by either measuring the external current ($RC \ll T$) or the voltage drop across the electrodes ($RC \gg T$), where T is the electron transit time. In the case $RC \ll T$, the external circuit current is

$$I(t) = \frac{eW_0}{d}\int_0^d n(z,t)\,{\rm d}z,$$

where d is the inter-electrode separation. The integral can be expressed in terms of TOF parameters, although some uncertainties arise in the treatment of boundary conditions. Although the parameter W_0 is usually referred to as the pulsed Townsend drift velocity, the experimental value of the drift velocity is obtained from the transit time of the swarm moving between the electrodes. Clearly this is the centre-of-mass velocity W (Tagashira 1981). In principle a measurement of W_0 could be made using the absolute value of $I(t)$, provided that an independent evaluation of the electron number density is made. As far as we are aware no experimental values of W_0 have been published.

4. Numerical Examples

Tagashira *et al.* (1977) have published TOF, SST and PT parameters in argon using a two-term Legendre series solution of the Boltzmann equation. For the smaller values of E/N [i.e. $\alpha_T = (1/n)(\partial n/\partial z)$ is small], it is expected that the SST parameters given in equations (10)–(12) could be approximated by the first two terms of the gradient expansion, so that

$$W_{\mathrm{SST}} \approx W_0 - \alpha_T D_\mu, \qquad (\nu_i)_{\mathrm{SST}} \approx \nu_{i0} - \alpha_T W_i.$$

Similarly for the TOF parameters we have

$$D_L \approx D_0 + D_\mu, \qquad W = W_0 + W_i,$$

and hence

$$W_{\mathrm{SST}} \approx W_0 - \alpha_T(D_L - D_0), \qquad (\nu_i)_{\mathrm{SST}} \approx \nu_{i0} - \alpha_T(W - W_0). \tag{14a, b}$$

By inserting the values for α_T and the TOF and PT parameters found by Tagashira *et al.* into equations (14), values for W_{SST} and $(\nu_i)_{\mathrm{SST}}$ are obtained ($N = 3{\cdot}54\times10^{16}$ cm^{-3} for their calculations). These are compared with the results of Tagashira *et al.* (1977) in Table 1, which also shows their values of ν_{i0}, W_0 and W.

Table 1. Values of transport parameters found in the present work and by Tagashira *et al.* (1977)

E/N	Present work $(\nu_i)_{\mathrm{SST}}$ (10^6 s^{-1})	Present work W_{SST} (10^6 cm s^{-1})	Tagashira *et al.* (1977) $(\nu_i)_{\mathrm{SST}}$ (10^6 s^{-1})	W_{SST} (10^6 cm s^{-1})	ν_{i0} (10^6 s^{-1})	W_0 (10^6 cm s^{-1})	W (10^6 cm s^{-1})
85	1·53	7·07	1·53	7·09	1·64	6·86	7·34
141	6·61	11·1	6·67	11·2	7·55	10·6	12·0
212	17·6	16·0	17·8	16·3	21·2	15·1	17·9
283	33·1	20·6	33·8	21·1	41·5	19·5	23·9
424	76·1	29·5	78·2	30·6	102	28·3	36·5
566	130	37·8	137	40·0	187	37·3	50·5

The agreement is excellent at the lower values of E/N, while the discrepancies at higher E/N are expected to be caused by the neglect of higher order terms in the derivation of equations (14).

5. Conclusions

Although the results presented in this work are not new, the relationship between the SST, TOF and PT parameters has not been clearly stated in the literature. In particular these quantities represent different groupings of the constant coefficients defined in equation (5) (additional coefficients are required for a three-dimensional swarm and there are modifications to the definitions for a multi-term Legendre expansion of the distribution function). The terminology of SST and PT parameters is confusing since only TOF parameters or related quantities are obtained from these experiments. In the presentation of theoretical calculations we believe that it would be more instructive to publish coefficients such as those defined in equation (5) together with TOF parameters. This would be of assistance in the numerical modelling of discharge development and give a better intuitive grasp of swarm behaviour.

The spatial variation of the energy distribution in either TOF or SST experiments can be observed more directly from the relative intensities of spectral lines originating from different excited states of the gas molecules. Using single photon counting techniques we have detected spatial variations in the relative intensities of spectral lines for a three-dimensional SST experiment using a localized electron source. The local excitation rate (for the jth excited state) can be written as

$$n(\boldsymbol{r},t)\,v_{\mathrm{ex}}^{(j)}(\boldsymbol{r},t) = V_1\,N\int_0^{\infty} Q_{\mathrm{ex}}^{(j)}\,\varepsilon^{\frac{1}{2}} f_0(\boldsymbol{r},\varepsilon,t)\,\mathrm{d}\varepsilon\,,$$

and after using a gradient expansion for $f_0(\boldsymbol{r},\varepsilon,t)$ this gives

$$n(\boldsymbol{r},t)\,v_{\mathrm{ex}}^{(j)}(\boldsymbol{r},t) = n(\boldsymbol{r},t)\,v_{\mathrm{ex0}}^{(j)} - \boldsymbol{V}_{\mathrm{ex}}^{(j)}\,.\,\nabla n + \dots .$$

Since this experimental procedure appears to be the most promising for investigating the internal structure of a swarm it is suggested that theoretical studies should include the evaluation of quantities such as $v_{\mathrm{ex0}}^{(j)}$ and $\boldsymbol{V}_{\mathrm{ex}}^{(j)}$. Comparison of these parameters with experiment would greatly assist in the evaluation of collision cross sections from swarm measurements at high E/N.

The present treatment of electron transport and rate coefficients does not include a detailed discussion of boundary conditions in finite systems. It is known (Kumar *et al.* 1980) that the simple boundary condition $n(\boldsymbol{r}) = 0$ can lead to significant errors in some circumstances and this limitation should be considered in applying transport theory to bounded discharges.

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