Momentum-dependent annihilation rate for positrons in metals

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Results are presented of a calculation for the partial annihilation rate of a thermalized positron with electrons in a metal as a function of the initial electron momentum. The method used has previously been successfully applied to the calculation of total annihilation rates. Recent work has shown that the agreement with experimental total rates is even better than was originally claimed. In this method the positron-electron wave function is calculated self-consistently using an effective interaction which includes strong interaction effects from no more than one highly correlated screening electron at any instant of time. Within the formalism self-energy insertions must be retained. The partial rates are more sensitive to these insertions than are the total rates. The calculated partial annihilation rates in the electron gas at metallic densities are all monotonically increasing functions of the initial electron momentum up to the Fermi surface. For lithium the partial rate is in good agreement with experimental data.

I. INTRODUCTION

In a previous paper[1] a scheme was developed for calculating the correlated electron wave function at a positron site in metals. The annihilation rate of positrons in metals is directly related to this quantity. Another quantity which can be measured is the partial annihilation rate for positrons. This is defined as the annihilation rate with electrons of one particular specified momentum. The momentum of a thermalized positron at room temperature is negligible compared with the Fermi momentum of conduction electrons at metallic densities, so the partial rates can be obtained by combining positron lifetime data with angular correlation data of the emitted γ rays. Although in practice experimental difficulties mean that the available data remains somewhat imprecise, the functional dependence of the partial rate on momentum is still a stringent test of any many-body theory of positron annihilation. In particular, the measured partial rates are all increasing functions of momentum up to the Fermi surface. In Ref. 1 the total rates were calculated directly without reference to the partial rates. We report here the result for the momentum-dependent partial annihilation rates using the same method. In particular, using this method we are able to obtain for the first time accurate partial rates for densities below $r_s = 4$.

The partial annihilation rate can be formally related to the correlated-positron–many-electron wave function. In our approach this wave function is calculated using an effective interaction which includes in a self-consistent manner the strong nonlinear screening effects from no more than one highly correlated screening electron at any instant of time. Since this one additional electron could equally well interact strongly with only the positron or only the electron, thus giving significant self-energy contributions, self-energy insertions on the intermediate propagators between interactions must be included.

The electron wave functions are approximately antisymmetrized using the analogy of a recoilless impurity. We emphasize that recoil is only neglected in so far as antisymmetrization is concerned and is not equivalent, as has been claimed,[2] to a total neglect of the antisymmetrizing Pauli projection operator. We further emphasize that in constructing the effective electron-positron interaction prior to antisymmetrizing, the recoil of the positron is included in the energy denominators. In Ref. 1 we selected this approximate antisymmetrization procedure for the electrons, only because we had already shown that the conventionally used Bethe-Goldstone equation[3] is not applicable to this problem. Briefly, the positron is an impurity which destroys the homogeneity of the electron gas, so that even at great distances from the positron, where its potential is completely screened out, the exact single electron wave functions will be phase shifted away from the unperturbed plane-wave states. In any practical calculation, the Bethe-Goldstone equation must use an unperturbed plane-wave basis, and so incorrectly predicts a zero scattering phase shift for positive momentum states lying below the Fermi level. This particularly leads to problems in the density range below $r_s = 4$ where the total annihilation rates calculated using the Bethe-Goldstone equation are known to diverge. The cause of this divergence was shown in Ref. 1 to be directly attributable to the Bethe-Goldstone equation's neglect of scattering into the Fermi sea of plane waves. The ladder approach when carried through correctly continues to yield reliable results in this low-density re-

gion for both the total rates and also the partial rates.

In Ref. 1 the calculational scheme was successfully used to predict the annihilation rate for simple metals spanning the entire range of conduction-electron densities. We should make two comments concerning the comparison in Ref. 1 of the calculated rates and the experimentally measured rates. Firstly, it has been subsequently pointed out[4] that a quoted experimental point at 5.0 nsec^{-1} for Al was measured using an incorrectly annealed sample, and should therefore be disregarded. Secondly, it was stated that in comparing the two results, a 20% core electron contribution should be subtracted from the experimental points. This was based on Carbotte and Salvadori's[5] estimates of the annihilation rate with core electrons for aluminium and sodium. However, a recent calculation has shown that the figure of 20% is a gross overestimate,[6] and that a more correct value for the core contributions is more than an order of magnitude smaller. We conclude from this that the theoretical curve and the experimental points in Ref. 1 should be compared directly. The parameter-free agreement between theory and experiment over a wide range of densities is remarkable. We also note that this theory alone predicts the correct low-density limit for the rate. The claim by Bhattacharyya and Singwi[7] to have overcome the divergence problem around $r_s=6$ was premature since their calculated rate for $r_s>4$ depends strongly on an undetermined parameter which is adjusted until the calculated rate at $r_s=6$ agrees with experiment. This is questionable on two grounds, (i) because the term with the parameter in it contains other factors which only become appreciable at densities below $r_s=4$ so that the rates for $r_s<4$ are insensitive to the parameter anyway, and (ii) because the rate calculated in this way drops to zero for $r_s \geq 7$ instead of approaching the positronium limit.

In Sec. II we recall the definition of the momentum dependent partial annihilation rate and outline our formalism for calculating it. In Sec. III we discuss our theory's self-energy insertions more fully, and in Sec. IV we present and discuss the results.

II. FORMALISM

The total annihilation rate for positrons in a uniform electron gas is given by

$$R=\lambda g_{+-}(r=0) \ , \tag{1}$$

where $g_{+-}(r)$ is the positron-electron correlation function and $\lambda=12/r_s^3$ nsec^{-1}. The quantity r_s is the usual density parameter of the unperturbed electron gas. $g_{+-}(r=0)$ can be formally expressed in terms of the full antisymmetric wave function of the many-particle system, consisting of N electrons at positions $\vec{r}_1, \vec{r}_2, \ldots, \vec{r}_N$ and the positron at $\vec{r}_p$:

$$g_{+-}(r=0)=\frac{1}{\Omega^{N-1}}\int d^3r_2 \cdots d^3r_N |\psi(\vec{r}_p, \vec{r}_p, \vec{r}_2, \ldots, \vec{r}_N)|^2 \ , \tag{2}$$

where Ω is the integration volume. With the approximations used in Ref. 1, Eq. (2) simplifies to

$$g_{+-}(r=0)=\sum_{|\vec{p}_j|<k_F} |\langle \psi^{(0,j)}|\, \vec{r}=0\rangle|^2 \ , \tag{3}$$

where k_F is the Fermi momentum, and $\psi^{(0,j)}(\vec{q})$ is the solution of a Lippmann-Schwinger integral equation for the correlated electron-positron pair. The initial momentum of the thermalized positron is zero and the initial momentum of the electron is $\vec{p}_j$. After multiple scatterings with the positron, the final momentum of the correlated electron is $\vec{q}$. Within the same approximations, the wave function $\psi^{(0,j)}(\vec{q})$ also gives us the partial annihilation rate $R(\vec{q})$, formally defined as[8]

$$R(\vec{q})=\frac{\lambda}{\Omega^N}\int d^3r_2 \cdots d^3r_N \left| \int d^3r_p e^{-i\vec{q}\cdot\vec{r}_p} \times \psi(\vec{r}_p, \vec{r}_p, \vec{r}_2, \ldots, \vec{r}_N)\right|^2 . \tag{4}$$

The total rate can be obtained from Eq. (4) after an integration over $\vec{q}$, but it can be more easily determined directly from Eq. (2) without first calculating the partial rates.

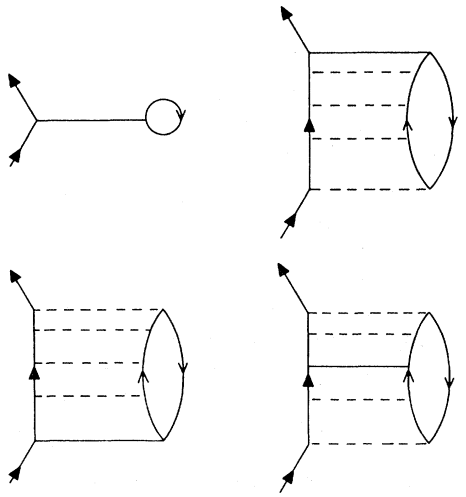

FIG. 1. Schematic representation of the contributions to the momentum-dependent partial annihilation rate from any infinite ladder series of terms. In each term the left most propagator with the solid arrow is the positron. The dotted horizontal lines represent the interaction which builds up the ladder sum. The solid horizontal line represents the appropriate density-density operator.

The wave function $\psi^{(0,j)}(\vec{q})$ is the solution of an infinite ladder sum. The four contributions to $R(\vec{q})$ for such a wave function are schematically shown in Fig. 1.[9] The horizontal dotted lines represent the effective interaction building up the ladder terms, while the solid horizontal line represents the appropriate density-density operator. The total contribution to $R(\vec{q})$ is then

$$R(\vec{q}) = \lambda \sum_{|\vec{q}|>k_F} \sum_{|\vec{p}_j|<k_F} \sum_{|\vec{p}_j-\vec{q}|\neq 0} \left| 1 + \psi^{(0,j)}(\vec{q}) \frac{1}{\epsilon_e^{(p)}(\vec{q}) + \epsilon_p^{(p)}(\vec{p}_j - \vec{q}) - \epsilon_e^{(h)}(\vec{p}_j) - \epsilon_p^{(h)}(0)} \right|^2 . \tag{5}$$

$\epsilon^{(p)}$ are particle energies, $\epsilon^{(h)}$ are hole energies and their subscripts e and p refer to electron or positron. It is necessary to distinguish between particle and hole energies, since the self-energy insertions are different for the two cases.

One task then is to determine the wave function $\psi^{(0,j)}(\vec{q})$, which we carry out using the same calculational scheme as in Ref. 1. We recall this wave function is related in a simple way to the effective two-particle interaction between the positron and electron. Retaining only terms in which no more than one additional electron is excited at any instant, we may carry out the infinite sum obtaining a Lippmann-Schwinger integral equation for the effective interaction

$$\langle \vec{q}\vec{K}|t_{ep}(E)|\vec{k}_0\vec{K}\rangle = \langle \vec{q}\vec{K}|V_{\text{eff}}(E)|\vec{k}_0\vec{K}\rangle + \int \frac{d^3p}{(2\pi)^3} \langle \vec{q}\vec{K}|V_{\text{eff}}(E)|\vec{p}\vec{K}\rangle \{1/[E - \epsilon_e^{(p)}(\tfrac{1}{2}\vec{K}+\vec{p}) - \epsilon_p^{(p)}(\tfrac{1}{2}\vec{K}-\vec{p})]\} \langle \vec{p}\vec{K}|t_{ep}(E)|\vec{k}_0\vec{K}\rangle . \tag{6}$$

Here $|\vec{q}\vec{K}\rangle$ is a positron-electron plane-wave state with relative momentum $\vec{q}$ and center-of-mass momentum $\vec{K}$. In the initial state the positron momentum is zero so that $\vec{K} = (2\vec{k}_0) = \vec{p}_j$, the initial momentum of the electron. E is the initial energy of the pair, equal to $\epsilon_e^{(h)}(\vec{K})$, since the zero momentum positron self-energy is vanishingly small on account of the low density of positrons. On the first iteration V_{eff} is simply the linear screened interaction from no more than one screening electron at any instant,

$$\langle \vec{q}K|V_{\text{eff}}(E)|\vec{p}K\rangle = V(|\vec{q}-\vec{p}|)\left[1 + \frac{2\Pi_0^{\text{TDA}}(|\vec{q}-\vec{p}|,E')\,V(|\vec{q}-\vec{p}|)}{1-\Pi_0^{\text{TDA}}(|\vec{q}-\vec{p}|,E')\,V(|\vec{q}-\vec{p}|)}\right] , \tag{7}$$

where V is the bare Coulomb interaction and Π_0^{TDA} is the forward part of the Lindhard function Π_0 without the sum over spins. Since particle self-energy insertions cannot overlap with V_{eff},

$$E' = \tfrac{1}{2}p^2 + \tfrac{1}{2}q^2 - E . \tag{8}$$

On subsequent iterations $V_{\text{eff}}^{(1)}$ includes the effect on screening of the nonlinear buildup of screening electron density around the positron. Each bare Coulomb interaction in Eq. (7) is replaced with the appropriate ladder sum of interactions. The positron-screening electron bare interaction V is replaced by t_{ep} as calculated in the previous iteration. Each electron-electron interaction should also be replaced by a ladder sum of unscreened Coulomb interactions, but in practice this modification only marginally affects the final results. The full expression for $V_{\text{eff}}^{(1)}$ is given in Ref. 1.

When the iteration procedure has converged, the resultant self-consistent interaction t_{ep} leads directly to the correlated electron-positron wave function through the relation

$$\psi^{(0,j)}(\vec{q}) = [1/(E-q^2)]\langle \vec{q}\vec{K}|t_{ep}(E)|\vec{K}\vec{k}_0\rangle . \tag{9}$$

III. SELF-ENERGIES

In our calculational scheme we must include self-energy insertions on the intermediate particle propagators between interactions, since the one screening electron could have equally well interacted strongly with only the positron or only the electron thereby leading to self-energy contributions. Terms with two self-energy insertions overlapping in time are excluded since they involve the strong excitation of more than one additional electron at some instant of time. Similarly, vertex corrections in which the retarded interaction overlaps with self-energy insertions must be excluded. The permitted self-energy insertions on the intermediate particle propagators are all a well-defined distance off the energy shell. Thus in Fig. 2 the second-order self-energy insertion in the intermediate electron line is

$$\Sigma_e^{(p)(2)}\left(\frac{\vec{K}}{2}+\vec{q},\omega\right)=\frac{1}{(2\pi)^4}\int d^3k'\int d\omega' e^{-i0^+\omega'}V(|\vec{k}'|)\Pi_0(\vec{k}',\omega')V(|\vec{k}'|)G_e^{(0)}\left(\frac{\vec{K}}{2}+\vec{q}+\vec{k}',\omega-\omega'\right), \tag{10}$$

where $G_e^{(0)}$ is the free-electron Green's function and

$$\omega=\epsilon_e^{(p)}\left(\frac{\vec{K}}{2}+\vec{q}\right)+\tfrac{1}{2}q^2-\epsilon_e^{(h)}(\vec{K}) \ .$$

Here the term $\frac{1}{2}q^2$ appears rather than $\epsilon_p^{(p)}(\vec{q})$, since the self-energy insertions in the intermediate positron propagator cannot overlap with $\Sigma_e^{(p)}$.

Strictly, each self-energy insertion in the intermediate particle lines should be calculated using the same infinite ladder sums as for the other correlated terms. In practice, we argue[1] that for small $\vec{k}$, the random-phase approximation should provide a good approximation for each self-energy insertion. For large $\vec{k}$ self-energy effects should be small, because the uncertainty principle restricts the time available for the original virtual excitation of momentum $\vec{k}$ to produce additional virtual excitations in self-energy-type processes.[10] Since the random-phase approximation insertion itself goes quite rapidly to zero for $|\vec{k}|>1.5k_F$, we use it to approximate self-energy effects for all $\vec{k}$, regarding it as an interpolating function between the high- and low-$|\vec{k}|$ limits.

Self-energy insertions on hole lines do not enter in the calculation in the same way as insertions on particle lines. Firstly, there is no restriction on them overlapping in time with additional electron excitations. Secondly, since only particle-particle scattering is to be included, a hole self-energy insertion corresponds to a quite different looking term from the corresponding particle insertion (see Fig. 3). This implies that an insertion on a hole line must be taken on the energy shell, even though it occurs on an internal line. Referring to Fig. 3, the reason is that the lifetime of the insertion, $t_{\lambda'}-t_\lambda$, is not dependent on the other times in the main part of the diagram. In contrast, the lifetime of the particle insertion in Fig. 2, $t_{\lambda'}-t_\lambda$, is necessarily less than the duration of the main positron-electron excitation t_2-t_1.

It is straightforward to perform the integrations over the time variables in Fig. 3. Retaining only the kinetic energies on the propagators shown, the resultant energy denominators for Fig. 3 are

$$\frac{1}{T(\vec{a})+T(\vec{b})-T(\vec{K})-T(\vec{c})}\left(\frac{1}{T\left(\frac{\vec{K}}{2}+\vec{q}\right)+T(\vec{q})-T(\vec{k})}\right)^2\frac{1}{T(\vec{d})+T\left(\frac{\vec{K}}{2}+\vec{q}\right)-T(\vec{f})-T(\vec{K})}, \tag{11}$$

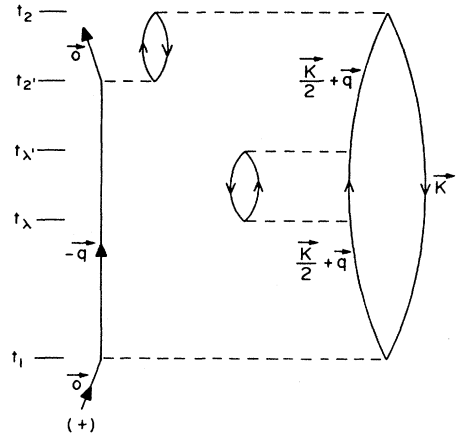

FIG. 2. Second-order self-energy insertion in the correlated-electron propagator. This term is permitted since the additional excited electron does not overlap in time with the excited screening electron. Relevant time labels are shown. Since the insertion is necessarily restricted to lie between the times t_1 and $t_{2'}$, it must be evaluated off the energy shell.

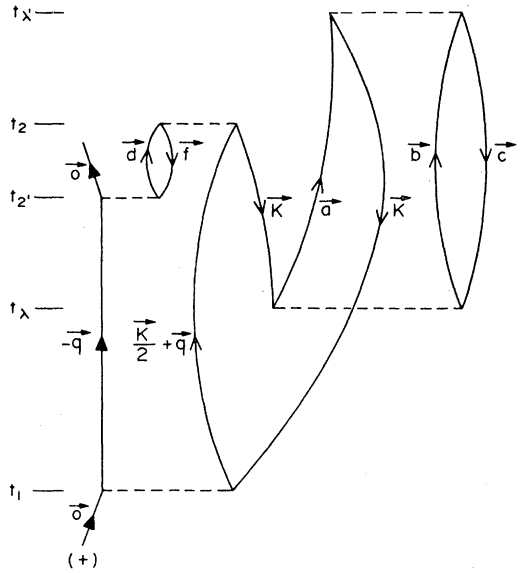

FIG. 3. Second-order self-energy insertion in the internal electron-hole propagator. This is to be compared with the corresponding particle insertion in Fig. 2. Because the insertion is not restricted to lie between the time t_1 and t_2, it "decouples" from the main part of the diagram and it must be evaluated on the energy shell.

where the kinetic energy $T(\vec{k}) = \frac{1}{2}|\vec{k}|^2$. But this is the same result as we would have obtained by directly evaluating the self-energy insertion on the energy shell. This simple argument can be generalized in a straightforward manner to all orders in the perturbation series.

Inclusion of self-energy insertions lowers the total annihilation rates by about 10%.[1] They affect the shape of the partial annihilation rate curves $R(\vec{q})$ even more significantly, lowering the rates for small $\vec{q}$ and pushing up the rates as $\vec{q}$ increases towards the Fermi momentum.

IV. RESULTS AND CONCLUSION

A calculation was carried out for the momentum-dependent partial annihilation rate for a positron in an electron gas at metallic densities. The procedure for calculating the nonlinear screening of the positron-electron interaction self-consistently, satisfactorily converged after one iteration, and the resultant self-consistent interaction t_{ep} led directly to $\psi^{(0,j)}(\vec{q})$ through Eq. (9).

We then used Eq. (5) to compute the partial rate $R(\vec{q})$. In Fig. 4 the solid lines show our calculated $R(\vec{q})$ for three electron gas densities. The partial annihilation rates at metallic densities are all monotonically increasing functions of the initial electron momentum $\vec{q}$ up to the Fermi surface. $r_s = 3$ corresponds to the conduction electron density in lithium.

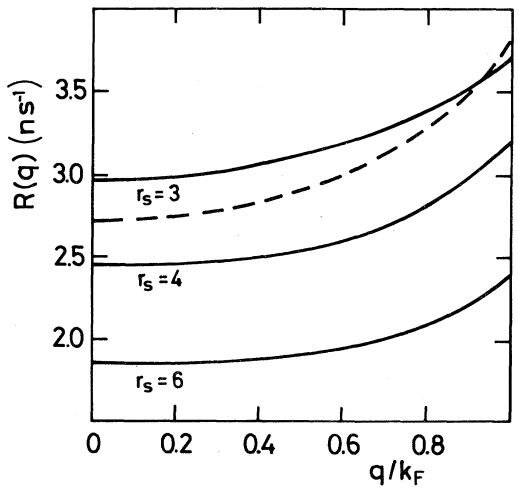

FIG. 4. Partial annihilation rates as a function of momentum. The three solid curves are, from the top, for electron gas densities of $r_s = 3$, 4, and 6, respectively. The dashed curve is the corresponding result for $r_s = 3$ taken from Ref. 3. (The rates in Ref. 3 cannot be accurately computed for densities below $r_s = 4$.) Note the monotonic increase of the partial rates as a function of q.

The dotted line shows $R(\vec{q})$ calculated by Kahana[3] for $r_s = 3$. The shape of this curve is somewhat steeper and lies significantly below our corresponding solid curve, this being a reflection of the fact that our calculated total rate for lithium lies about 10% above Kahana's value.[1]

Assuming the positron is at rest we can calculate the number of γ rays possessing a total momentum q_z, in some fixed direction, by numerically integrating our $R(\vec{q})$ over an appropriate plane intersection of the Fermi sea. We obtain the distribution function

$$P(q_z) = \int_{q_z}^{k_F} dq' q' R(q') \quad . \tag{12}$$

If $R(q')$ is roughly constant over the sea, Eq. (12) will give the familiar inverted parabola distribution observed for many metals.

Kim and Stewart[11] have managed to differentiate their coincident photon counting rate numerically to obtain a plot of $qR(q)$. This experimental quantity provides a much more sensitive test of any theory than the integrated result of Eq. (12). In Fig. 5 we show their reported data for lithium at 80 K together with our calculated $qR(q)$ curve for $r_s = 3$. Kim and

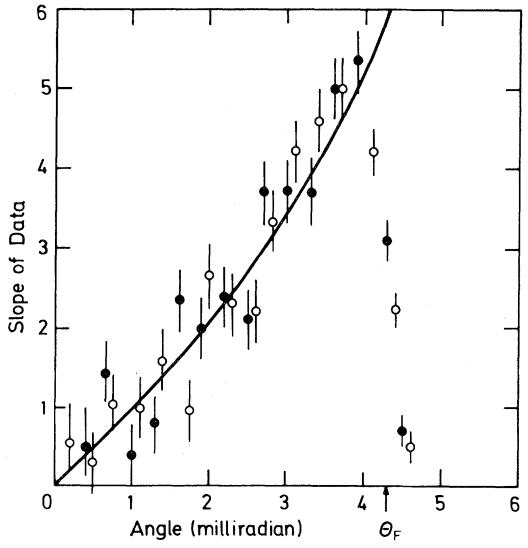

FIG. 5. Experimental data points, taken from Ref. 11, of the slope of the coincident photon counting rate for positrons in lithium at 80 K. The optical resolution of the measuring instrument has a full width at half maximum of 0.32 mrad. The angle θ_F corresponds to an electron with momentum k_F. This slope is directly related to the momentum-dependent partial rate and provides a more stringent test of any theoretical prediction than the integrated distribution function. The solid curve shows the corresponding $qR(q)$ from the present calculation. The vertical scale for the experimental data has been interpreted such that the data is consistent with the experimentally observed total rate for lithium.

Stewart's scale on the derivative axis is not reported and we have chosen the scale so their partial rate when integrated over $\vec{q}$, gives the observed total rate for lithium, 3.4 $nsec^{-1}$. The data points are not consistent with any single fitted curve, and our curve shows satisfactory agreement with their points up to an angle corresponding to about $0.9k_F$. Our calculation does not predict the apparent rapid falloff in $qR(q)$ as q approaches the Fermi surface, but the optical resolution of the instrument used in this experiment had a full width at half maximum of 0.32 mrad and this causes a smearing of the data. Thus a sizeable part of the falloff in this region arises from contributions to $qR(q)$ outside the Fermi surface, where $qR(q)$ is known to be small. Our curve also shows the steepening that is characteristic of a partial rate which is increasing across the Fermi sea.[3]

Our conclusion is that the present calculation, which avoids the serious objections associated with using the Bethe-Goldstone equation for this problem,[1,2] not only gives remarkable agreement with the observed total rates—much better agreement than was claimed in Ref. 1—but it also leads to partial rates which show the well-known increase with momentum across the Fermi sea and which are consistent with the best available experimental data for lithium.

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