This article can be cited before page numbers have been issued, to do this please use: V. Podolskiy, P. Ginzburg, B. Wells and A. Zayats, *Faraday Discuss.*, 2014, DOI: 10.1039/C4FD00186A.

View Article Online
DOI: 10.1039/C4FD00186A

Light emission in nonlocal plasmonic metamaterials

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DOI: 10.1039/b000000x [DO NOT ALTER/DELETE THIS TEXT]

We present analytical and computational studies of light emission in nonlocal metamaterials formed by arrays of aligned plasmonic nanowires. We demonstrate that emission lifetime in these composites is a complex function of geometrical and material parameters of the system that cannot be reduced to "trivial" hyperbolic or elliptical dispersion topology of a homogenised metamaterial. In particular, our studies suggest that Purcell factor can often be maximized when the composite operates in elliptic regime, with strong radiation coupling to additional TM-polarized mode supported by the nonlocal composite, in contrast to the accepted "hyperbolicity related" enhancement.

1 Introduction

Metamaterials, composites designed from dissimilar components of subwavelength size, offer new avenues of controlling light generation, propagation, and absorption [1,2]. Recently, plasmonic nanowire arrays emerged as a flexible platform for confinement and manipulation of optical pulses at deep subwavelength scale, along with applications in negative refraction, sensing, and nonlinear optics [3]. It was demonstrated, experimentally and numerically, that optical response of these composites is strongly affected by spatial dispersion of effective permittivity – the dependence of components of the permittivity tensor on a wavevector [4]. Analytical description of optical nonlocality in nanowire arrays has recently been developed [5]. In particular, it was shown that optical nonlocality yields additional wave, extra TM-polarized optical mode, that has hyperbolic-like properties at frequency ranges where main TM wave has elliptic dispersion. In this work, we use the nonlocal effective medium theory to analyse emission of point dipoles in nanowire media and show that nonlocality significantly affects light emission in metamaterials, enhancing photonic density of states in elliptic regime, in contrast with previous theoretical proposals [6] that predict that Purcell enhancement is maximized in hyperbolic regime. We relate the apparent contradiction to the excitation of plasmonic modes in metamaterial constituents, missed in the local effective medium theory, and argue that the nonlocal effective medium theory provides a more adequate description of underlying physics.

The manuscript is organized as follows – we first describe the modal structure of the nanowire metamaterial and provide the relationship between geometrical parameters of the metamaterial, its spatially dispersive permittivity, and dispersion of optical modes in the composite. The developed nonlocal effective medium theory (EMT) allows solving boundary condition problem for plane wave incidence. We then employ the plane wave decomposition of a dipolar emmiter for calculation of spontaneous emission rates via classical-quantum correspondence and the formalism

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of the radiation reaction force.

2 Nonlocal optical response of nanowire media

2.1 Optical modes supported by nanowire metamaterial

Plasmonic nanowire arrays represent collections of aligned metallic wires (with permittivity ϵ_i and average radius r) embedded into dielectric host (permittivity ϵ_h) with mean centre-to-centre separation a. Nanowire metamaterials typically operate in the effective medium regime where $r < a \ll \lambda_0$ with $\lambda_0 = 2\pi c/\omega$ being free-space wavelength, ω being angular frequency of optical radiation, and c being speed of light in vacuum. In this regime optical properties of the composite may be well described by averaged geometric parameters (concentration $p = \pi r^2/a^2$; [here $p \ll 1$] and, in case of nonlocal effective medium theory, shape of the unit cell) and are weakly affected by the details of wire distributions (variations of wire radius or unit cell size across the composite). In this work we follow the approach introduced in Ref. [5] and consider nanowire metamaterials having square unit cell (see Fig.1).

As follows from symmetry considerations, nanowire metamaterials have uniaxial anisotropic response with an optical axis parallel to the wires ($\hat{z}$ axis in this work). In Cartesian coordinates used in this work, the effective permittivity tensor is diagonal, with components $\epsilon_{xx} = \epsilon_{yy} = \epsilon_\perp$, $\epsilon_{zz} \neq \epsilon_\perp$. In quasistatic limit, the Maxwell-Garnett effective medium theory predicts the following relation between effective permittivity ϵ^{mg}, permittivities of components of the composite, and the geometrical parameters of the metamaterial:

$$\epsilon_\perp^{mg} = \epsilon_h \frac{(1+p)\epsilon_i + (1-p)\epsilon_h}{(1+p)\epsilon_h + (1-p)\epsilon_i};\ \epsilon_{zz}^{mg} = p\epsilon_i + (1-p)\epsilon_h \qquad (1)$$

As any uniaxial material, the modes supported by the infinite nanowire metamaterial can be characterized according to their polarization. The modes of the first kind, often referred to as ordinary waves or transverse-electric (TE) polarized waves, have their electric field in the xy plane. Propagation of these waves is not affected by the metamaterial anisotropy and is completely determined by $\epsilon_\perp$ component of the permittivity tensor.

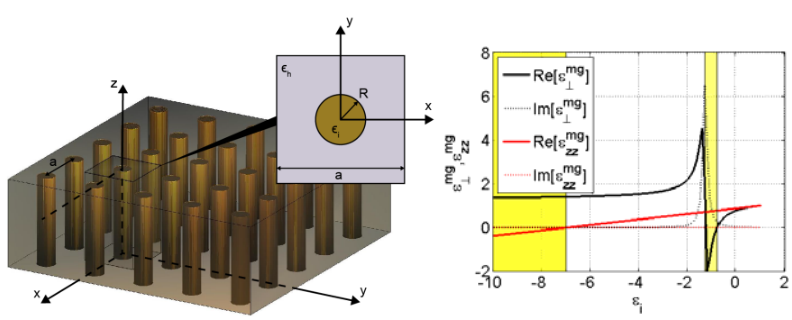

Fig.1 Geometry of nanowire composite (left) and the local effective medium parameters (right); $Im(\epsilon_i) = 0.1$; shaded areas represent spectral range where metamaterial operates in hyperbolic regime; when $\epsilon_i \gtrsim -1$ metamaterial is local; additional wave exists for $\epsilon_i \lesssim -1$; this wave propagates inside metamaterial for $-7 \lesssim \epsilon_i \lesssim -1$ (main wave has elliptical dispersion); when $\epsilon_i \lesssim -7$, longitudinal wave exponentially decays along the wires, while the main wave has hyperbolic dispersion

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In contrast to this behaviour, propagation of the waves that have non-vanishing component of electric field in the $\hat{z}$ direction is (strongly) affected by material anisotropy. Depending on relationship between ϵ_i, ϵ_h, and p, these extraordinary (TM polarized) modes have elliptic (when $\epsilon_\perp^{mg} \cdot \epsilon_{zz}^{mg} > 0$) or hyperbolic (when $\epsilon_\perp^{mg} \cdot \epsilon_{zz}^{mg} < 0$) dispersion (see Fig.1).

In addition to these transverse electromagnetic waves, a plasmonic nanowire metamaterial may support propagation of longitudinal electromagnetic waves that represent coupled cylindrical surface plasmon (CSP) waves propagating along the wires. As shown in Ref.[5], dispersion of these waves can be related to the solution of eigenvalue problem with eigenvalues and eigenvectors representing propagating constants (k_z^l) and sets of amplitudes, respectively, describing contributions of individual cylindrical waves propagating in the nanowires of the composite to the coupled CSP modes. These CSP modes do not exist when wires are dielectric ($\epsilon_i > 0$), they propagate in the plasmonic elliptic regime ($\epsilon_{zz}^{mg} > 0, \epsilon_i < -\epsilon_h$); and exponentially decay along the wires in the hyperbolic regime ($\epsilon_{zz}^{mg} < 0$). In the limit of PEC wires ($\epsilon_i \to -\infty$), propagation constant of longitudinal wave approaches the limit $k_z^l \to n_\infty^l \omega/c$.

From the effective medium standpoint, the same dispersion relation can be written as $\epsilon_{zz}(k_z, \omega/c) = 0$ with

$$\epsilon_{zz}(k_z) = \xi\left(k_z^2 - k_z^{l^2}\right)\frac{c^2}{\omega^2};\;\; \xi = p\frac{\epsilon_i+\epsilon_h}{\epsilon_h-\left(n_\infty^l\right)^2}. \qquad (2)$$

Note that spatial dispersion, explicit dependence of permittivity on wavevector, is a necessary condition for existence of the longitudinal electromagnetic wave.

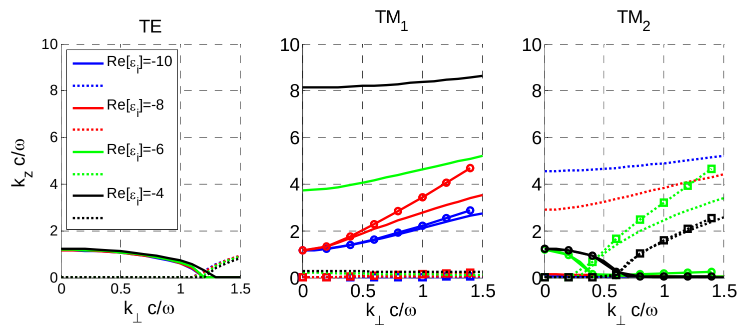

Fig.2 Dispersion of the three waves supported by metamaterial for different permittivities of the nanowires: (solid line) $Re[k_z c/\omega]$, (dashed line) $Im[k_z c/\omega]$, (symbols) dispersion of "main" TM-polarized mode according to local EMT. All other parameters as in Fig. 1.

Although transverse electromagnetic waves that enter nanowire material at normal incidence cannot couple to a longitudinal mode, light that obliquely enters metamaterial can couple into both longitudinal mode and transverse modes. In a strict effective medium approach, light propagating inside metamaterial should be represented as a linear combination of three modes. The first of these modes is an ordinary TE wave, described above, whilst the other two represent a mix of the "local" transverse TM and "nonlocal" longitudinal waves. Dispersion of these two waves and the relative contributions of "local" and "nonlocal" components are given by the coupled-oscillator-like expression,

$$\left(k_z^2 - k_z^{l^2}\right)\left(k_z^2 - \epsilon_\perp^{mg}\frac{\omega^2}{c^2}\right) = -\frac{\epsilon_\perp^{mg}}{\xi}\frac{\omega^2}{c^2}k_x^2. \qquad (3)$$

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To better illustrate our results, in this work we fix operating wavelength and geometric parameters of the metamaterial, permittivity of dielectric host medium, and vary permittivity of wire inclusions. For the set of parameters used in this work ($r = 20nm, a = 100nm, \epsilon_h = 1, \lambda_0 = 1.5\mu m$), epsilon near-zero (ENZ) transition occurs at $\epsilon_i \simeq -7$ with elliptic regime realized for $\epsilon_i > -7$, and hyperbolic regime realized for $\epsilon_i < -7$ (see Fig.1 for predictions of local EMT). Dispersions of the three modes propagating inside nanowire materials are shown in Fig.2. Note that in regime when the main TM mode is elliptic, the additional wave has hyperbolic-like dispesrion.

2.2 Transmission and reflection of light into nonlocal nanowire metamaterials

To understand transmission and reflection of optical waves by nanowire metamaterials with finite thickness, solutions of Maxwell equations in each of the bulk layers are represented as linear combination of (plane) waves that differ by their polarization and direction (towards the interface or away from the interface; see Fig.3). The amplitudes of the waves in the neighbouring regions are then related to each other via boundary conditions. This relationship is then used to calculate the amplitudes of the waves propagating away from the interface as a function of the amplitudes of the waves propagating towards the interface. Such linear relationship between the amplitudes can be conveniently represented in terms of the scattering (S-) matrix.

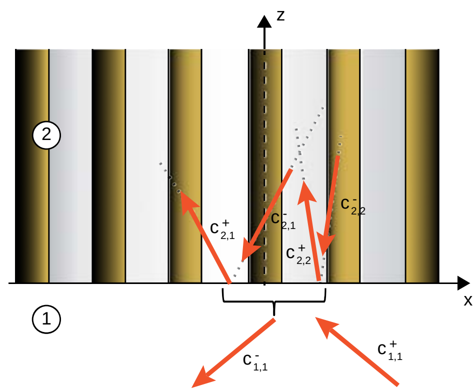

Fig.3 Schematic of light reflection and transmission at the interface between local and nonlocal media

In conventional, local materials, it suffices to enforce the continuity of tangential components of electric and magnetic fields to calculate S-matrix of the interface. In nonlocal metamaterials, existence of additional (TM-polarized) mode requires introduction of additional boundary conditions (ABCs). Here, we will follow the recipe of Ref.[5] and use the continuity of tangential electric field, the continuity of normal electric displacement, and the continuity of the first moment of normal electric displacement (product of $D(x) \cdot \exp 2\pi i\, x/a$, averaged over the unit cell) to calculate the S-matrix of each interface in the system.

3 Light emission in nanowire metamaterials

In this work we follow the well-developed technique that relates the enhancement in

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decay rate of dipole to the regular part of the field generated by this dipole at the origin. Explicitly,

$$\frac{\Gamma}{\Gamma_0} \simeq \frac{3}{2}\frac{Im(\vec{E}\cdot\vec{P})}{\omega^3|P|^2} \tag{4}$$

where $\vec{E}$ represents the field generated by the point dipole $\vec{P}$. The main objective of this work is to analyse enhancement of decay rate in plasmonic nanowire composites and to relate this enhancement to spectral properties of the composites. It is worth noting that placing a dipole directly inside ideal hyperbolic or realistic (lossy) material leads to unphysical singularities [7,8]. Various real or virtual cavity approaches were suggested to address the impact of the surrounding media on a host emitter[9]. To address these key issues, in this work we remove singularity inside the hyperbolic metamaterial by limiting the wavevector spectrum used in calculations of the field $\vec{E}$ in Eq.(4) and by employing the following three-step strategy that allows us to address the issue of material absorption.

First, we calculate emission of the point dipole inside hypothetical, lossless, metamaterial. Predictions of both local and nonlocal effective medium theories are analysed, with two TM-polarized modes in nonlocal metamaterials taken as two competing decay channels.

Second, emission of the point 3D dipole positioned in the small (vacuum-filled) slit surrounded by two infinite slabs of hypothetical lossless metamaterial is analysed. Combined with first step, these calculations allow us to estimate geometry-dependent local-field-correction effects. Local field correction is the frequently employed technique to account for the arrangement of the emitter and the host matrix. It is worth noting, that the geometry of the artificially created cavity could affect the spontaneous emission rate; here we use the slit geometry, approaching its physical dimensions to zero thickness.

Finally, we calculate emission of the point dipole positioned in the small vacuum slit surrounded by lossy metamaterials and use the local field correction, estimated above, to calculate modulation of dipole lifetime due to metamaterial.

3.1 Plane wave expansion of Green's function in homogeneous material

To calculate modification of a dipole lifetime, we represent Green's function of anisotropic metamaterial as a set of plane waves, and use Eq.(4) to calculate modification of dipole lifetime. Since we consider emission inside anisotropic material, the field generated by the dipole needs to be separated into TE and TM-polarized components.

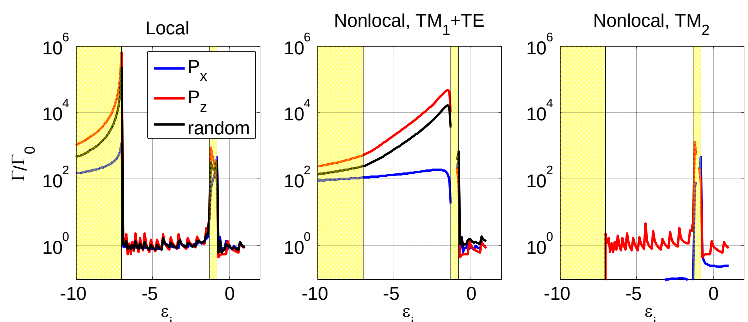

Fig. 4 Enhancement of decay rate in infinite lossless metamaterials (lines of different colours represent different orientation of the dipole);

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It can be explicitly shown [10] that the field of the $\hat{z}$ polarized dipole is TM-polarized, with the regular part of $E_z^{TM} = iP\epsilon_\perp \int_0^{k^{max}} k^3 dk/(k_z \epsilon_{zz})$, where the integration parameter k represents in-plane component of the wavevector. In this work, we take into account the fact that any effective medium theory fails in the limit when $k \simeq \pi/a$. We therefore constrain the integrals above to the range of wavevectors inside the first Brillouin zone of our metamaterial, $k < k^{max} = \pi/a$.

On the other hand, $\hat{x}$ polarized dipole emits both TE- and TM-polarized waves, with amplitudes yielding the plane-wave expansion of the field: $E_x^{TM} = iP \int_0^{k^{max}} k_z k\, dk/(2\epsilon_\perp)$, and $E_x^{TE} = iP \int_0^{k^{max}} \omega^2 k\, dk/(2c^2 k_z)$. Due to uniaxial symmetry of the problem, y-component of electric field generated by a $\hat{y}$-polarized dipole is identical to E_x generated by $\hat{x}$-polarized dipole.

3.2 Emission in lossless metamaterials and local field correction analysis

We begin by analysing the emission of a point dipole positioned inside an infinite material described by the local effective medium parameters given by Eq.(1), keeping in mind the high-wavevector cut-off due to finite unit cell size. The results of these calculations are shown in Fig.4. It is seen that, in agreement with previous studies [6], the local effective medium theory suggests that enhancement of photonic density of states can be linked to (i) plasmon resonance in the wires (corresponding to $\epsilon_i \simeq -1$) and (ii) broadband spectral range starting from ENZ ($\epsilon_i \simeq -7$) and continuing through the hyperbolic regime of metamaterial dispersion ($\epsilon_i < -7$). Interestingly, ENZ regime seems to provide maximum reduction of lifetime.

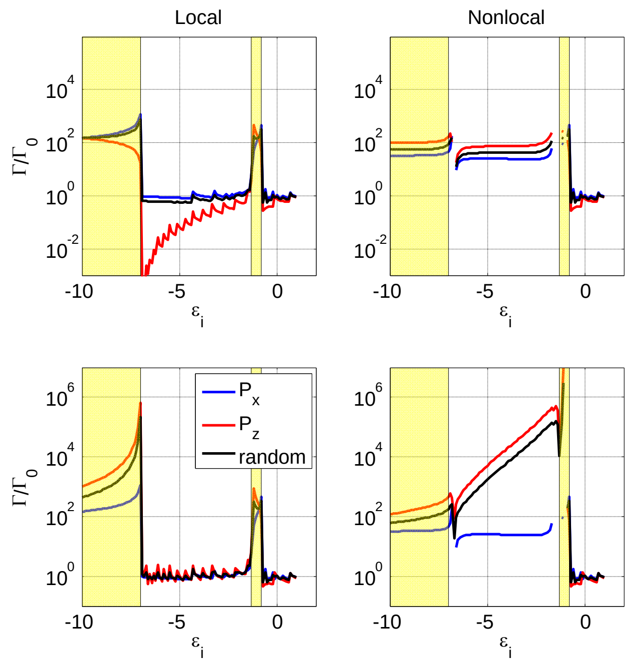

Fig.5 Emission of the point dipole positioned in the vacuum slit inside the lossless nanowire metamaterial; top and bottom rows correspond, respectively, to results of calculations that are not and are accounted for local field corrections

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Calculations of the lifetime modifications have been repeated for a situation when a point dipole is positioned inside the homogeneous material described by the nonlocal EMT model [Eqs.(2,3)]. In this case, we assume that the dipole can independently emit into two TM-polarized channels. As seen in Fig.4, predictions of the nonlocal EMT drastically differ from predictions of its local counterpart. The main difference, dramatic enhancement of the decay rate in the elliptic regime can be traced to the existence of an additional electromagnetic mode, originating from collective excitation of CSPs in nanowire composite. Note that hyperbolic-like CSP-rich mode (mode TM_1 in Fig.2) dominates emission across both elliptic and hyperbolic regimes. Also note that nonlocality removes the singularity associated with ENZ regime (at $\epsilon_i \simeq -7$).

To analyse emission of dipole in realistic, lossy, metamaterials, it becomes necessary to place the dipole inside substantially small cavity carved in the homogeneous material. In this work, we assume that the cavity takes a shape of a small planar slit, oriented perpendicular to the wires. Emission of the point dipole is calculated according to plane-wave expansion, described above, incorporated into transfer-matrix formalism, as suggested in Ref.[11].

For substantially small slit size (below $\sim 10\,nm$), enhancement of decay rate becomes independent of the slit size due to high-wavevector k_{max} cut-off described earlier. Furthermore, for a local metamaterial it becomes possible to eliminate local field correction effects by renormalizing emission of $\hat{z}$-polarized dipole by $1/\left(\epsilon_{zz}^{mg}\right)^2$ (Fig.5). Similarly, renormalization of emission of $\hat{z}$-polarized dipole by $1/\epsilon_{zz}(k)^2$ positioned inside the slit carved in the nonlocal metamaterial also yields results that qualitatively agree with total decay rate calculated for a dipole positioned inside the nonlocal medium (exact nature of the resonance predicted by the nonlocal EMT in ENZ region will be addressed in our future work). Therefore, we use the above normalization to take into account local field correction effects.

3.3 Effect of finite material absorption

Finally, we calculated a decay rate of the dipole positioned in the slit between two slabs of lossy nanowire metamaterial with $Im(\epsilon_i) = 0.1$ (Fig.6). As expected, presence of material absorption does not fundamentally alter the dynamics of the emission. In general, increase in $Im(\epsilon_i)$ leads to further enhancement of decay rate thoughout the “plasmonic” spectral range ($\epsilon_i < -\epsilon_h$).

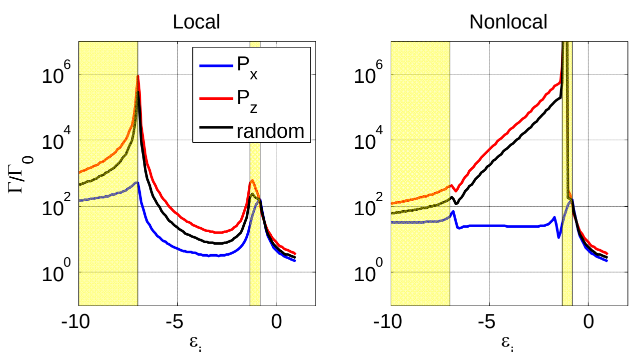

Fig.6 Enhancement of decay rate in metamaterials with finite lossess; the plots take into account local field correction; $Im(\epsilon_i) = 0.1$; see Fig.1 for effective medium parameters

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4 Discussion

It is clear that predictions local and nonlocal effective medium theories regarding enhancement of a decay rate for a dipole emitting inside nanowire metamaterial disagree with each other. It therefore reasonable to ask which (if any) theory accurately predicts the response of the dipole in the real metamaterial. The fundamental reason for the disagreement between the predictions of the two effective medium theories lies in the existence of additional wave in nonlocal EMT. The existence of additional wave has been validated in both full-wave numerical solutions of Maxwell equations in nanowire composite and in experimental studies[4,5]. It is therefore reasonable to assume that the nonlocal EMT adequately describes emission inside the composite. Our main results can be interpreted in the following intuitive manner: *the fundamental change in the density of optical states in a nanowire medium is associated not with the onset of hyperbolicity, but rather with onset of plasmonic response in nanowires.*

Similar effect is expected in layered materials with low fill-fraction where layer's transition to plasmonic response is spectrally separated from composite's transition to hyperbolicity. It should be also mentioned that in realistic nanowire composites enhancement of decay rate will be affected by standing wave Fabry-Perot type resonances.

5 Conclusions

We have analysed enhancement of a decay rate of the dipole emitting inside the plasmonic nanowire composite. Predictions of the nonlocal effective medium response, previously demonstrated to adequately describe refraction in nanowire composites [5] suggest significant enhancement of decay rate across the spectral range where nanowire are plasmonic, and in particular across the spectral range where the composite as the whole exhibits elliptic response.

Acknowledgements

This research was supported by US Army Research Office (Grant # W911NF-12-1-0533), EPSRC (UK), and the ERC iPLASMM project (321268). A.Z. acknowledges support from the Royal Society and the Wolfson Foundation.

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