Comment on "Normalization of quasinormal modes in leaky optical cavities and plasmonic resonators"

E. A. Muljarov and W. Langbein

School of Physics and Astronomy, Cardiff University, Cardiff CF24 3AA, United Kingdom
(Received 23 February 2016; published 10 July 2017)

Recently, Kristensen, Ge, and Hughes [Phys. Rev. A **92**, 053810 (2015)] have compared three different methods for normalization of quasinormal modes in open optical systems and concluded that they all provide the same result. We show here that this conclusion is incorrect and illustrate that the normalization of P. T. Kristensen *et al.*, Opt. Lett. **37**, 1649 (2012), is divergent for any optical mode having a finite quality factor, and that the Silver-Müller radiation condition is not fulfilled for quasinormal modes.

In a recent paper [1], Kristensen *et al.* have considered three different normalizations of quasinormal modes: (i) the normalization given in [2], which is a generalized version of the work by Leung *et al.* [3] and thus called here Leung-Kristensen (*LK*), (ii) the normalization introduced in [4], which is analytically *exact*, and (iii) the normalization suggested in [5], based on perfectly matched layers (*PML*). Kristensen *et al.* concluded that all three normalizations provide the same result. We show in this Comment that (i) the LK normalization is divergent, and therefore ill defined. A regularized variant of the LK normalization, put forward in [1], is not suited for numerically determined resonant states (RSs), and (ii) the claimed equivalence of LK and PML normalizations is incorrect since the Silver-Müller radiation condition used in the argumentation is not valid for RSs. We emphasize that the spectral representation of the Green's function is converging to the exact Green's function, and the resulting spectral summation for the Purcell factor is converging to the exact Purcell factor, only if the exact mode normalization is used. It is therefore of utmost importance to have a well-defined expression for the exact normalization which can be evaluated efficiently. We clarify here that the LK normalization does not have such properties.

The LK normalization, Eq. (5) of [1], for an optical system surrounded by vacuum is defined by an infinite-volume limit

$$N_{\mathrm{LK}}^{\infty} = \lim_{V \to \infty} N_{\mathrm{LK}} \tag{1}$$

of the normalization

$$N_{\mathrm{LK}} = \int_V \varepsilon(\mathbf{r}) \mathbf{E}^2(\mathbf{r}) dV + \frac{i}{2k} \oint_{S_V} \mathbf{E}^2(\mathbf{r}) dS, \tag{2}$$

calculated over the finite volume V and its surface S_V, using the electric field $\mathbf{E}(\mathbf{r})$ and the wave vector k of the quasinormal mode, which we call here *resonant state*, adopting its original name in the literature [6]. Let us assume for now that the volume is a sphere of radius R with the system in its center.

We first show that N_{LK} diverges for $R \to \infty$, so that the LK normalization N_{LK}^{∞} mathematically does not exist. The dependence of N_{LK} on R was evaluated in [1] by expanding $\mathbf{E}(\mathbf{r})$ into vector spherical harmonics, with the spherical harmonics $Y_{lm}(\theta,\varphi)$ and Hankel functions of first kind $h_l(z)$ as basis (here l is the orbital quantum number). Since k is complex for any RS having a finite quality factor (Q factor) $Q = |\mathrm{Re}(k)/[2\,\mathrm{Im}(k)]|$, the argument of $h_l(z)$ is

also complex: $z = kR$. The limiting form of $i^{l+1} h_l(z) \to e^{iz}/z$ given in Eq. (9) of [1] neglects diverging contributions, since the exact form is given by

$$i^{l+1} h_l(z) = \frac{e^{iz}}{z} P_l(\xi), \tag{3}$$

where

$$P_l(\xi) = \sum_{m=0}^{l} \frac{(l+m)!}{(l-m)!m!} \xi^m, \quad \xi = \frac{1}{-2iz}. \tag{4}$$

Now, $P_l(\xi)$ is a polynomial of order l, and all resulting terms of Eq. (3) diverge for complex z, owing to the exponentially large factor e^{iz}. Consequently, Eq. (10) in [1], based on Eq. (9) and stating that $\partial_R \hat{I}_l^r(R) = 0$, is incorrect, and should read instead

$$
\partial_R \hat{I}_l^r(R) = R^2 h_l(z) \left[h_l(z) + i h_l'(z) + i \frac{h_l(z)}{z} \right]
$$
$$
= \frac{h_l^2(kR)}{2k^2} \frac{P_l'(\xi)}{P_l(\xi)}. \tag{5}
$$

In particular, $P_l'(0)/P_l(0) = l(l+1)$, and thus $\partial_R \hat{I}_l^r(R) = 0$ holds only for $l = 0$. However, electromagnetic modes with $l = 0$ do not exist in finite three-dimensional optical systems. Therefore, in general, $\partial_R \hat{I}_l^r(R) \to \infty$ for $R \to \infty$. For example, considering $l = 1$ we find

$$\partial_R \hat{I}_1^r(R) = \frac{e^{2ikR}}{k^4 R^2} \left(1 + \frac{i}{kR} \right). \tag{6}$$

The authors of [1] write: "In practice, direct application of Eq. (5) leads to an integral that seems to quickly converge towards a finite value, but in fact oscillates about this value with an amplitude that eventually starts to grow (exponentially) with the distance, albeit slowly compared to the length scales in typical calculations. This was noted in Ref. [5], where the oscillations were observed only for the cavity with the lowest quality factor ($Q \approx 16$)." In the cited reference (Ref. [2] in this Comment), we find: "For very low-Q cavities, however, the convergence is nontrivial due to the exponential divergence of the modes that may cause the inner product to oscillate around the proper value as a function of calculation domain size," and otherwise "quick convergence" is claimed. The residual $f_{\mathrm{LK}}^{\mathrm{res}}(R)$ of the LK normalization, which is given in Eq. (11)

of [1] diverges—its precise form is

$$f_{\text{LK}}^{\text{res}}(R) = \frac{R^3}{2}\left[h_l^2(z) - h_{l-1}(z)h_{l+1}(z) + \frac{i}{z}h_l^2(z)\right]$$

$$= \frac{e^{2ikR}}{k^5 R^2}Q_{2l-2}(\xi), \tag{7}$$

where $Q_n(\xi)$ is an nth order polynomial of $\xi = (-2ikR)^{-1}$, with the leading term at small ξ (i.e., at large R) given by $Q_{2l-2}(0) = -i(-1)^{l+1}l(l+1)/2$; see [7] for more details. Therefore, $N_{\text{LK}} \to \infty$ as $R \to \infty$.

The authors of [1] describe this divergence as follows: "Thus, while Eqs. (9) and (10) appear to be formally correct also for complex arguments, the limit $R \to \infty$ in practice leads to a position dependent phase difference between the Hankel function and its limiting form, which makes the limit nontrivial to perform along the real axis." We note that (i) there is no difference between formalism and practice in mathematical limits; (ii) the limit $V \to \infty$ is defined for real volumes, and thus real R; (iii) the limit of N_{LK} along the real axis of R is not "nontrivial"; it simply does not exist due to the divergence.

We show in Figs. 1–3 the R dependence of N_{LK} for RSs of a dielectric sphere of radius a with high and low Q factors, and for the fundamental plasmonic RS of a gold sphere. All RS fields used have been normalized using the exact normalization, having analytical expressions [7,8]. We commence using a RS with a Q factor of about 35 (similar to the RS illustrated in Fig. 3 of [1]), the $l = 7$ transverse electric (TE) whispering gallery mode (WGM) of a dielectric sphere with refractive index $n_r = 2$ in vacuum. Figure 1 is formatted similar to Fig. 3 of [1], showing in blue the R region of convergence (spiralling in), and in red the R region of divergence (spiralling out) of N_{LK} in the complex plane. We note that the spiralling out region is not shown in Fig. 3 of [1].

One could argue that for high-Q modes, the LK normalization can be sufficiently accurate, as the error reaches 10^{-3} at $R \approx 10a$ in the present example. One could even refine this result by evaluating the center of the spiral, as suggested in [1]. However, one has to keep in mind that simulating the required extended spatial domain in numerical calculations is computationally costly. On the other hand, evaluating the LK normalization close to the system leads to significant errors due to the slow $1/R^2$ dependence of the residual term Eq. (7), as is clearly shown by the blue line in Figs. 1(b) and 1(d). The LK normalization used for high-Q RSs is thus at least inconvenient, due to the large computational domain required to obtain sufficient accuracy. More discussion and data are given in the Appendices C and G of [7].

A RS with a low Q factor of about 1 in the same dielectric sphere, a TE $l = 7$ leaky mode, is used in Fig. 2. We see that N_{LK} starts close to zero at $R = a$ and then spirals out in the complex plane. This results in an initial error of about 100%, increasing to 40 000% at $R = 2a$, prohibiting to extract a value for the LK normalization.

Finally, we show in Fig. 3 the LK normalization of the fundamental surface plasmon mode of a nanoplasmonic resonator—a gold sphere in vacuum, 200 nm in diameter, also used in [5]. This mode has a Q factor of about 1.3. There is an initial decrease of the error from 200% down to about 10%, followed by an exponential divergence. A single loop in the
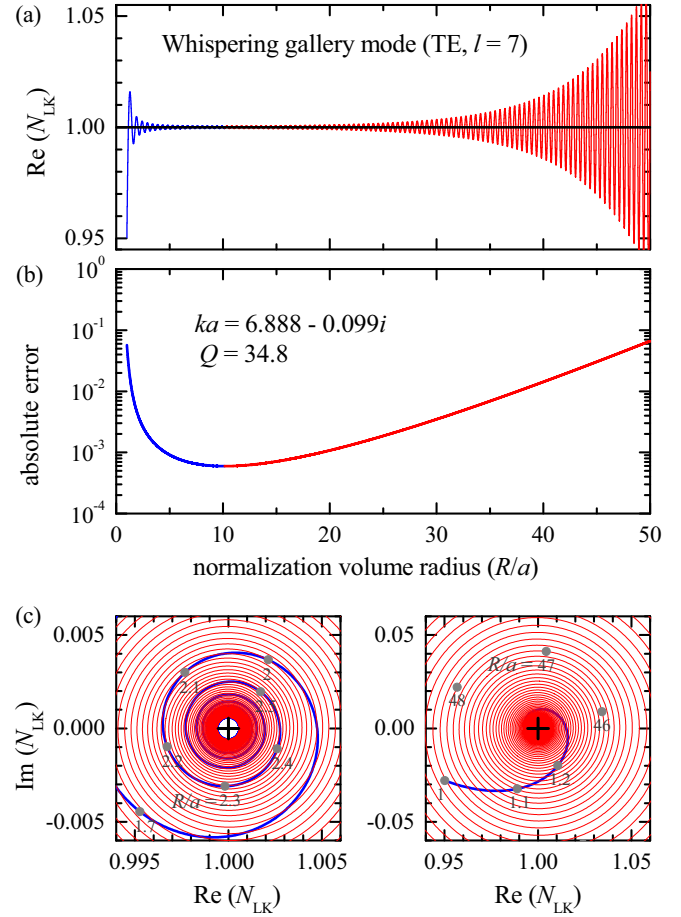

FIG. 1. LK normalization N_{LK} (a),(c)–(d) and its absolute error $|N_{\text{LK}} - 1|$ (b) as a function of the radius R of the spherical volume, for a TE $l = 7$ WGM of a dielectric sphere of refractive index $n_r = 2$ and radius a, in vacuum. The wave vector of the WGM is $ka = 6.888 - 0.099i$, corresponding to $Q = 34.8$. Blue (red) color shows the region of error decreasing (increasing) with R. The exact normalization is shown by a black line (a) and a black cross (c)–(d).

complex plane is observed, circling the correct normalization. The minimum of the error is observed at about $R = 3a$, thus requiring a much larger computational domain than the system size. A reliable extraction of the RS normalization from N_{LK} in this case is questionable.

A regularized version of the LK normalization suggested in [1] is based on an analytic continuation of the electric field into the complex plane of R. While [1] fails to define the contour of integration used, we can find in [9] that the contour can involve taking the limit of $R \to -\infty + i\epsilon$. For this to be applied, the fields of RSs have to be known analytically. This regularization is thus not suited for numerically determined RSs. We emphasize that this "regularized" LK normalization is a different quantity compared to the divergent LK normalization defined by Eqs. (1) and (2), the one that was actually used in [2] and in numerous followup publications of the same group, including the numerical examples of [1]. In particular, Eq. (27) of [1] is incorrect, because its left-hand side is the LK normalization taking infinite value, while its right-hand side is the regularized LK normalization taking a finite value.
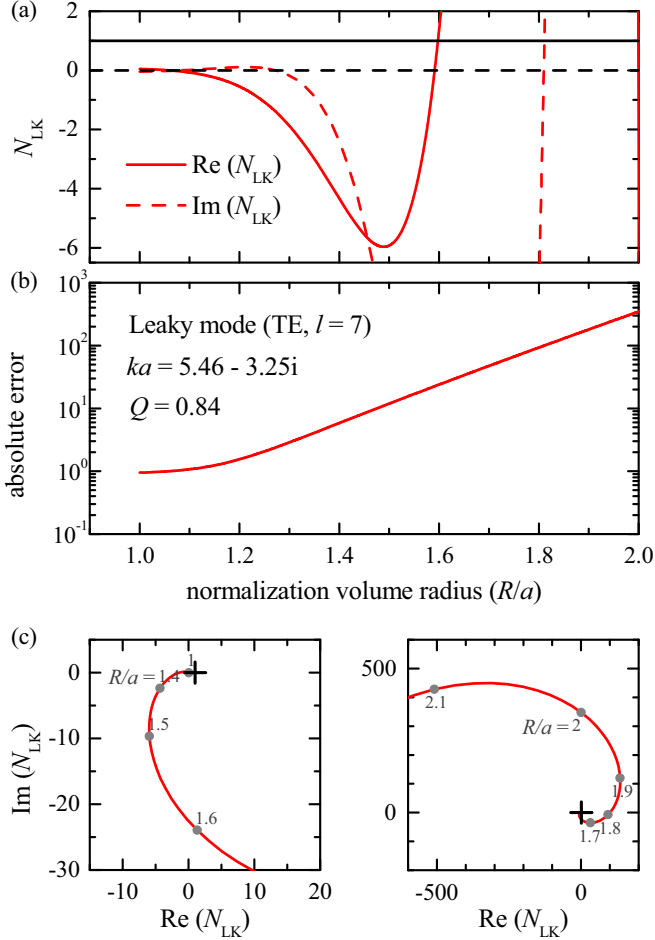
FIG. 2. As in Fig. 1 but for a leaky mode with the wave vector $ka = 5.46 - 3.25i$, corresponding to $Q = 0.84$.
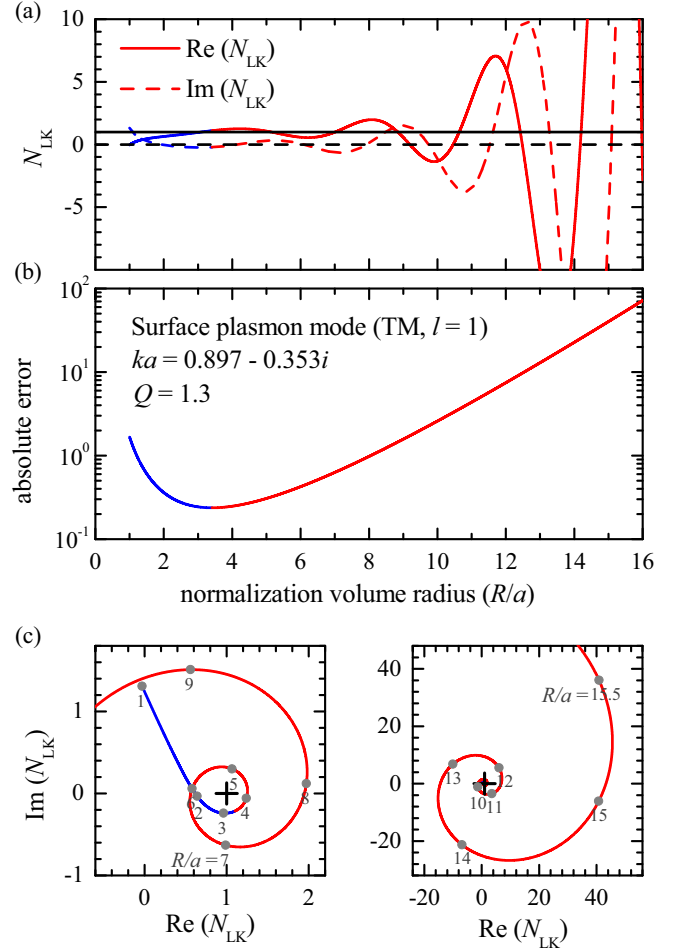
FIG. 3. As in Fig. 1 but for a surface plasmon $l = 1$ transverse magnetic (TM) mode in a gold sphere in vacuum. The mode wave vector is $ka = 0.897 - 0.353i$, corresponding to $Q = 1.27$. The radius of the sphere is $a = 100$ nm, and the gold permittivity was treated using a Drude model [5].

The exact normalization [4] is independent of V and differs from the LK normalization only by the surface term. To understand the physical difference between the surface terms, we consider a small piece ΔS of the surface of integration and assume for simplicity that the local electric field of the RS has the form of a plane wave $\mathbf{E} = \mathbf{E}_0 e^{i\mathbf{k}\cdot\mathbf{r}}$ propagating in the direction of $\mathbf{k}$, with $\mathbf{k}^2 = k^2$ and a constant amplitude $\mathbf{E}_0$. Then, after simple algebra, we find that the selected part of the surface integral in the exact normalization is given by

$$\frac{i}{2k^2} \int_{\Delta S} (\mathbf{k} \cdot \hat{\mathbf{n}}) E^2 dS, \qquad (8)$$

where $\hat{\mathbf{n}}$ is the surface normal, while for the LK normalization the corresponding part is

$$\frac{i}{2k} \int_{\Delta S} E^2 dS. \qquad (9)$$

This shows that the LK surface term assumes that the propagation direction of the field is always normal to the surface, while the exact normalization takes the actual propagation direction into account. The two terms are equal only if $\hat{\mathbf{n}} \parallel \mathbf{k}$ over the whole surface, which is not possible in electrodynamics due to the vectorial nature of the electromagnetic field.

The implicit assumption of normal outward propagation makes the LK normalization not only diverging for $V \to \infty$, but also depending on the surface shape. Note that the shape of S_V in the LK normalization is not restricted to spherical surfaces, and a cuboid was actually used in one of the examples shown in [2] and [1]. However, since the surface term in N_{LK} is independent of the surface normal, it changes proportionally to the surface area when the shape of the surface is modified. For example, by "roughening" the spherical surface to $R(\varphi) = R_0(1 + \epsilon \sin m\varphi)$, the surface integral scales as $\sqrt{1 + \alpha\epsilon^2 m^2}$, where α is a geometrical factor of order one, weakly dependent on the argument ϵm. As a result, N_{LK} can take arbitrary values, adjustable by the modulation amplitude ϵ and the spatial frequency m. At the same time, each piece of surface term in the exact normalization is proportional to the flux of $\mathbf{k}$, as clear from Eq. (8), and thus independent of the surface roughness.

Finally, we show that the claim in [1], that the LK normalization is equivalent to the PML normalization of [5], is incorrect. This should be clear considering that N_{LK} diverges, while the PML normalization is finite, as demonstrated in the

Supplemental Material of [5] for the RS shown in Fig. 3. The PML normalization uses a PML to convert the radiation losses into absorptive losses within the PML, such that the remaining radiation losses at the external border of the PML can be neglected.

The equivalence of the LK and PML normalization is shown in [1] analytically, using the Silver-Müller radiation condition. This condition states that the vector field

$$\mathbf{F} = \frac{\mathbf{r}}{r} \times \nabla \times \mathbf{E} + ik\mathbf{E} \tag{10}$$

vanishes at large distances from the optical system, i.e., $\mathbf{F} \to 0$ as $r \to \infty$. Here $\mathbf{E}$ is the electric field of a wave emitted from the system centered at the origin, with a wave vector k which is *real* and positive [10]. However, for a RS, k is typically *complex*, so that the Silver-Müller condition does not hold, and a divergence $\mathbf{F} \to \infty$ as $r \to \infty$ is found instead. To exemplify this, we take TE vector spherical harmonics, which can be used, along with their TM counterparts, for expansion of any mode of a finite system in the outside area. Their field can be written as

$$\mathbf{E} = -\mathbf{r} \times \nabla f, \quad \text{where } f(\mathbf{r}) = h_l(kr)Y_{lm}(\theta,\varphi), \tag{11}$$

so that

$$\mathbf{F} = \frac{\mathbf{r}}{r} \times [2 - ikr + (\mathbf{r} \cdot \nabla)]\nabla f$$

$$= \frac{h_l(kr)}{2ikr^2} \frac{P_l'(\xi)}{P_l(\xi)} \left(\mathbf{e}_\varphi \partial_\theta - \mathbf{e}_\theta \frac{\partial_\varphi}{\sin\theta} \right) Y_{lm}(\theta,\varphi), \tag{12}$$

in which $\mathbf{e}_\varphi$ and $\mathbf{e}_\theta$ are the unit vectors of the spherical coordinate system, and $\xi = (-2ikr)^{-1}$. We see that $\mathbf{F}$ diverges for $r \to \infty$ due to the exponentially growing factor in $h_l(kr)$ and the nonvanishing factor $P_l'(\xi)/P_l(\xi) \to l(l+1)$. In particular, the claimed equivalence of Eqs. (17) and (5) in [1], allowing the authors to obtain the LK normalization from the PML normalization, uses the property $\mathbf{F} \to 0$ for $r \to \infty$. This shows actually that the two normalizations differ by a term proportional to $\mathbf{F}$ which is diverging for $r \to \infty$, consistent with the fact that the LK normalization is diverging while the PML normalization is not.

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