This is a nice and well written paper, including some improvements of the literature on gravitational multipole moments, extending the Geroch-Hansen definition to stationary non-vacuum spacetimes, and its equivalence with Thorne's definition. I do recommend its publication in SciPost. However, I have several questions and comments, which hopefully help improving the paper before its publication.

1. In vaccum GR, the twist one-form $\boldsymbol{\omega}$ is defined geometrically and is not ambiguous. It is closed and allows for the definition of the twist scalar as $\boldsymbol{\omega} = d\omega$ [1]. However, in the presence of matter, $d\boldsymbol{\omega} \neq 0$ and it is suggested to add an improvement one-form $\boldsymbol{\omega}^I$ such that $d\boldsymbol{\omega}^{\mathrm{tot}} = 0$ where $\boldsymbol{\omega}^{\mathrm{tot}} = \boldsymbol{\omega} + \boldsymbol{\omega}^I$. Accordingly, a scalar can be found such that $d\omega = \boldsymbol{\omega}^{\mathrm{tot}}$. However, it is clear that the above construction is ambiguous: $\boldsymbol{\omega}^I \to \boldsymbol{\omega}^I + d\alpha$ does not affect the closedness of $\boldsymbol{\omega}^{\mathrm{tot}}$, however, it implies the ambiguity $\omega \to \omega + \alpha$ where α is an arbitrary smooth scalar. In particular, if ω is also smooth, we may choose $\alpha = -\omega$, which implies that the improved twist scalar is vanishing, which is a problem for the definition of spin multipole moments. I don't find a discussion of this ambiguity in the paper, in a way that leads to a unique definition of spin multipole moments in the generic non-vacuum stationary spacetimes. Looking at Sec. 3.1.2, while $W^{(2)}$ is uniquely defined in terms of matter stress tensor, $B^{(1)}$ suffers from similar ambiguity.

2. This paper assumes the case where the matter field is not isolated (if it is, then both Geroch-Hansen and Thorne methods already apply without modification). Now if matter is not isolated in any region of spacetime, then is there an argument why (14) should hold? Thorne writes down this equation for isolated (localized) sources (and this is stressed in his paper), where one solves vacuum Einstein equations in the exterior region, and the splitting of spacetime into near zone and far zone is meaningful. However, in the non-vacuum case, none of these arguments apply and it is not clear why (14) should hold. Note that the form (14) at linear order is derived from the multipole expansion of the Green's function $|x - x'|^{-1}$ in the region where $|x'| < |x|$, i.e. the observation point is farther. Again the inequality $|x'| < |x|$ is not applicable when the source extends all over the spacetime. I am not saying that (14) is wrong. I just don't see why it is true. It is nice if the author addresses this issue in the paper, or refers to the relevant literature.

3. The author resorts to the notion of asymptotic flatness several times, without being explicit what that means? Does it refer to a conformal compactification, or certain falloff conditions? At spatial infinity or null infinity? It would be appropriate to cite any references which discuss asymptotic flatness in the presence of non-localized matter.

4. I am not sure if this paper falsifies the claim of ref.[10]. In a stationary vacuum solution, it is possible to reconstruct the metric (up to diffeos) in terms of the two sets of mass and spin multipoles. The main question here is whether the same statement holds in the presence of non-isolated matter in Einstein equations. The paper actually assumes that it is true, by writing (14) in the generic situation, but it does not provide an argument to support this. From what I understand, what the paper does is to show that there is a procedure to define mass and spin multipoles in the generic case, but does not contain a discussion whether one needs additional multipoles for the bulk reconstruction or not. It would be nice if the author expands a bit on this and if needed rephrase the way [10] is cited.

5. I am not sympathetic with the first sentence of the paper. Multipoles are more related to the expansion of angular dependence into (spherical or STF) harmonics (irreducible reps of $SO(3)$), rather than to a radial expansion of the fields. For example, in (8.13) of Thorne, multipole moments are defined without using "asymptotic radial expansion" of the fields.

6. In (50), there is no discussion of the function $\mathcal{C}$. Why is that so? Is it free to have any growing r dependence asymptotically?

7. Remove duplicate "also" in the beginning of sec. 2.3.

8. Below (29), the index μ in $d\omega^{\mathrm{tot}}_\mu$ seems to be extra.

9. Correct typo (extra "a") above (48) and also below (51).

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[1] The author might want to consider using the above notation to distinguish the twist one form $\boldsymbol{\omega}$ from the twist scalar ω.

10. I don't understand the discussion at the end of Sec.3.3.2 regarding non-stationary metrics. Note that

$$\partial_i f(t - r) = -\frac{n_i}{r} \partial_t f$$

and therefore it again increases the power of $1/r$ in the expression. So, I don't see a difference between stationary and non-stationary in this regard.