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COMMENT

Comment on 'Semiclassical approximations in phase space with coherent states'

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Abstract

In two recent papers (Baranger *et al* 2001 *J. Phys. A: Math. Gen.* **34** 7227; 2002 *J. Phys. A: Math. Gen.* **35** 9493) co-authored by us, we mentioned the work of Dr Kenneth G Kay (Kay 1994 *J. Chem. Phys.* **100(6)** 4377; **100** 4432) in an unfavourable light. In this comment we correct this impression, as it was based on a misunderstanding of his work. The point at issue has been the derivation of the Herman–Kluk formula (Herman and Kluk 1984 *Chem. Phys.* **91** 27). We find Kay's derivation to be the most solid at the moment.

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In a recent paper [Bar01] with the above title, co-authored by us, and in our reply [Bar02] to a subsequent comment [Gro02] on it, we mentioned the work of Kay [Kay94a, Kay94b] in an unfavourable light. After several discussions with Kay, we now wish to correct this impression, as our description of his work was based on a misunderstanding of it.

The point at issue has been the validity of the Herman–Kluk [Her84] approximation (referred to in the following as HK) in providing an initial value integral expression for the propagation of wavefunctions in some sort of semiclassical limit. It is universally accepted that the HK expression gives very good results in many cases. For a number of specific problems, Kay himself has done numerical calculations [Kay94b] comparing it with other possible approximations, and he has found the HK method to be clearly superior. One of the points of our paper [Bar01], and the main point of our subsequent reply-to-comment [Bar02], was that the two existing microscopic derivations [Her84, Gro98] of HK both contain fatal mathematical errors. If one repeats these derivations, as we have done, without committing the errors, one obtains in one case a formula known as Heller's thawed Gaussian approximation (TGA) [Hel75], and in the other case a formula similar to Heller's and given in our paper [Bar01]. Both these formulae are quite different from HK. This leaves one facing a great mystery: how does the HK method, whose derivations are incorrect, manage to give better results than almost anything else people have tried? Clearly this problem needs to be solved and, with the help of Kay, we are trying to solve it. Meanwhile it has become clear to us that the most convincing derivation of HK at the moment is that given by Kay himself [Kay94a]. His derivation does not explain why HK is better than the other methods; one needs the numerical calculations [Kay94b] in order to see that. But at least it explains why HK is a possible method among others.

A very important aspect of this subject, and the source of our misunderstanding of Kay's papers, is the distinction that exists between a propagator based on a single classical trajectory and that expressed as an integral over many trajectories. HK is meant to be used as an integral expression over many trajectories; when its kernel is used as a one-trajectory propagator it performs very poorly. On the other hand, the TGA is a one-trajectory wave packet; as a kernel in an integral expression, it is at best mediocre. The reason for this huge difference between the two categories of propagators which would appear at first sight to perform the same function, is to be sought in the fact that the basis states used are not orthogonal, and therefore questions about unitarity do not have easy answers. Profound consequences follow from this, about which we hope to have more to say in the future.

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