Comments

Comments are short papers which criticize or correct papers of other authors previously published in the **Physical Review**. *Each Comment should state clearly to which paper it refers and must be accompanied by a brief abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Discrepancy in the temperature behavior of the modified impulse approximation and the third sum rule

G. Solt
Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland
(Received 24 May 1990; revised manuscript received 9 November 1990)

The discrepancy, found in a recent numerical study, in the temperature dependence of the cross-section formula resulting from a suggested modification of the impulse approximation is shown to originate in a spurious factor of $\simeq T^2$ present in the third central moment of the proposed dynamic structure factor.

In a recent paper Mayers *et al.*[1] consider the modification, proposed by Stringari,[2] of the impulse approximation (IA) to the incoherent dynamic structure factor $S(\kappa,\varepsilon)$ of neutron scattering. They find that this prescription, denoted by SIA in the following, "works well for two systems" at low temperatures and this, together with some general arguments, "provides strong support for its validity."

Their numerical comparison of the exact S, the standard impulse approximation S_{IA} and the proposed S_{SIA} in the case of a harmonic crystal proves, in any case, that the *temperature dependence* of the SIA formula is completely erroneous: S_{SIA} shows a markedly wrong behavior for all not too low temperatures $T \gtrsim \hbar\omega_D$ even at arbitrary large recoil energy $R=\hbar^2\kappa^2/2M$ [Figs. 2(a)–(c) in Ref. 1], whereas the "unmodified" S_{IA} for $R \gg \hbar\omega_D$ becomes a fairly good approximation to S at intermediate or high temperatures. (Here κ and ε are momentum and energy transferred to the scatterer and ω_D is Debye frequency; since IA has a sense only for the "deep inelastic" scattering $R > \hbar\omega_D$,[3,4] and SIA is meant[2] for the same domain, our discussion is limited to this range.) Noticing this failure, Mayers *et al.* define a "crossover temperature" $T_c \approx 0.3\hbar\omega_D$ below which SIA would be preferable to IA.

The aim of this Comment is to point out that, though SIA is *principally* wrong in its prediction for *S for all temperatures,* the error manifests itself most spectacularly in the *increasingly large asymmetry* of S_{SIA} as the temperature augments. This is demonstrated by the explicit T dependence of the skewness of S discussed below, explaining the numerical findings of Ref. 1.

Why and how SIA *must fail* to describe the variation of the asymmetry of S with T is immediately seen by looking at the third central moment s_3 of S, a general measure of its asymmetry about $\langle \varepsilon \rangle = R$. The known sum rule[5] for s_3 gives

$$s_3=\int_{-\infty}^{\infty}(\varepsilon-R)^3 S(\kappa,\varepsilon)d\varepsilon = R\hbar^2\langle \Delta V\rangle/3M \;, \tag{1}$$

where ΔV is the Laplacian of the (isotropic) potential binding the scattering nucleus of mass M in the condensed state; $\langle \Delta V\rangle = 3M\langle \omega^2\rangle$ is an "average force constant" for a harmonic system. The skewness, in energy units, is conveniently defined as $\varepsilon_s = s_3/s_2$, where $s_2=\frac{4}{3}R\langle E_{\text{kin}}\rangle$ is the second central moment.[5] (The dimensionless *coefficient of skewness* usually defined is $\alpha_3=\varepsilon_s/s_2^{1/2}$ which diminishes, in our case, with increasing R. We are interested here, however, in the behavior of S as a function of ε at a given κ and not as a function of the reduced variable $\varepsilon/s_2^{1/2}$.) By Eq. (1) and the quoted value of s_2 one has

$$\varepsilon_s = s_3/s_2 = \hbar^2\langle \Delta V\rangle/4M\langle E_{\text{kin}}\rangle \;. \tag{2}$$

Before comparing the T dependence of ε_s in Eq. (2) with that predicted by SIA, we look at a related quantity $\delta\varepsilon_0$, the shift, with respect to R, of the energy value ε_0, where S has its maximum. The relation between $\delta\varepsilon_0$ and skewness ε_s is found on the basis of the Nelkin-Parks expansion for S:[3,4]

$$S(\kappa,\varepsilon)=1/(2\pi^{1/2}\alpha)\exp^{-[(\varepsilon-R)/2\alpha]^2}\left[1+\sum_{n=3}^{\infty}\zeta_n(\kappa)H_n\left[\frac{(\varepsilon-R)}{2\alpha}\right]\right] , \tag{3}$$

where the first, exponential factor is $S_{\rm IA}$ for systems with Gaussian momentum distribution, and subsequent terms with even- and odd-parity Hermite polynomials H_n build up the symmetric, respectively antisymmetric components of S. Here $\alpha=(s_2/2)^{1/2}=(\frac{2}{3}R\langle E_{\rm kin}\rangle)^{1/2}$, and ζ_n's are algebraic expressions of the central moments s_n;[4] for example, $\zeta_3=2^{1/2}s_3/3s_2^{3/2}=\varepsilon_s/3\alpha$, which is $2^{1/2}/3$ times just the coefficient of skewness α_3 defined above. For a Gaussian system ζ_n's are, for n even, of the order of $\simeq 1/R$, and $\zeta_5,\zeta_7,\ldots$ contain, generally, terms with first and higher powers of $1/R^{1/2}$. For a system with an arbitrary, non-Gaussian momentum distribution a partial summation leading to $S_{\rm IA}$ can first be done,[4] and the ζ_n's in the remaining series have the above property. By equating the derivative of S in Eq. (3) to zero[4] and retaining only the leading antisymmetric term H_3 we find

$$\delta\varepsilon_0\equiv\varepsilon_0-R\simeq -s_3/2s_2=-\hbar^2\langle\Delta V\rangle/8M\langle E_{\rm kin}\rangle\ . \qquad (4)$$

To arrive at this result, $\delta\varepsilon_0/2\alpha<1$ has been assumed at the outset, which is indeed consistent with the solution Eq. (4), since $\delta\varepsilon_0$ is seen to be *independent* of R, as is ε_s in Eq. (2), and $\alpha\sim R^{1/2}$. The contribution of the omitted terms with $H_5,H_7,\ldots$ to $\delta\varepsilon_0$ is not easy to estimate in general,[6] when all of them effectively contain terms first order in $1/R^{1/2}$, but for a harmonic crystal this is not the case, the expansion Eq. (3) is asymptotic in powers of this parameter[3] so that Eq. (4) is seen to hold in $O(1/R^{1/2})$. Further, with $\langle\Delta V\rangle/3M=\langle\omega^2\rangle$ and $\langle E_{\rm kin}\rangle=\frac{3}{2}T(1+\hbar^2\langle\omega^2\rangle/12T^2-\cdots)$, we obtain

$$\delta\varepsilon_0\simeq-\varepsilon_s/2=-\hbar^2\langle\omega^2\rangle/4T+O(1/T^3) \qquad (5)$$

for a harmonic system.

The above expressions for ε_s and $\delta\varepsilon_0$ show how the asymmetry of S *disappears* with increasing T: that is why the symmetric $S_{\rm IA}$ can become *at all* a reasonably good approximation to S at higher temperatures (Fig. 2 of Ref. 1). By the proposed SIA prescription, however, the maximum occurs[2] at $\varepsilon_{0,\rm SIA}=R-\langle E_{\rm kin}\rangle$, and a direct calculation gives $s_{3,\rm SIA}\simeq\frac{8}{3}R\langle E_{\rm kin}\rangle^2$ in $O(1/R)$. Thus, since $\langle E_{\rm kin}\rangle\sim\frac{3}{2}T$ for $T/\hbar\omega_D\gtrsim 1$, both $s_{3,\rm SIA}$ and $\delta\varepsilon_{0,\rm SIA}$ contain, as compared to Eqs. (1) and (4), an *additional factor* $\sim T^2$, leading to an *increase* instead of a *reduction* of the skewness of $S_{\rm SIA}$ as T rises.

Although completely wrong in its temperature dependence, the SIA prescription might still work for $T=0$, i.e., for $T\ll\hbar\omega_D$, as proposed[2] and suggested also very recently.[7] To be sure, the symmetrical $S_{\rm IA}$ becomes, in this range, insufficient even for relatively large R's; in fact, an "antisymmetric" component (with respect to $\varepsilon=R$) of S was observed, at momentum transfers $\kappa\simeq 10$ Å^{-1}, for low temperature liquids like helium and neon,[8–10] so that the possibility to obtain new information on the dynamics of the scatterer via Eq. (2) or (4) arises.

Can this information be obtained by the SIA? As noted above, this prescription shifts the position of the maximum of S by $\delta\varepsilon_{0,\rm SIA}=-\langle E_{\rm kin}\rangle$, which is of the right *sign*, but it *does not correspond* to Eq. (4) [for example, for low temperatures $s_{3,\rm SIA}/s_3=3\langle\omega\rangle^2/2\langle\omega^2\rangle$ in $O(1/R)$ for the harmonic crystal]. On trying to describe width and skewness of S with a single parameter $\langle E_{\rm kin}\rangle$, the SIA necessarily requires $8M\langle E_{\rm kin}\rangle^2$ to stand for $\hbar^2\langle\Delta V\rangle$, a procedure with no justification even at low temperatures.[6]

On the other hand, lower-T high-R data are usually interpreted on the basis of Eq. (3), where the correction terms complementing $S_{\rm IA}$ insure that the sum rules, including Eq. (1), are *exactly* satisfied. On lowering T the ζ_n's increase and, for a general system, terms with $n\geq 4$ in Eq. (3) may be important indeed for any T. In spite of this practical difficulty, Eq. (3) represents a sound basis for data analysis even at low temperatures.[4,6,11]

In conclusion, $\varepsilon_s=s_3/s_2$ characterizing the skewness of S is proportional to the quantity $\hbar^2\langle\Delta V\rangle/M\langle E_{\rm kin}\rangle$; this determines in leading order also $\delta\varepsilon_0$ and varies for $T\gtrsim\hbar\omega_D$ as $1/T$ for a harmonic system, in contrast to $\varepsilon_{s,\rm SIA}$ or $\delta\varepsilon_{0,\rm SIA}$ which *increase linearly* with T. This gross violation of the third (and, for the matter, also the higher) sum rules by the SIA follows from its enforced one-parameter description of the dynamics of the scatterer, which amounts to the loss of the dynamical information contained in Eqs. (2) and (4).

[1] J. Mayers, C. Andreani, and G. Baciocco, Phys. Rev. B **39**, 2022 (1989).
[2] S. Stringari, Phys. Rev. B **35**, 2038 (1987).
[3] M. S. Nelkin and D. E. Parks, Phys. Rev. **119**, 1060 (1960).
[4] V. F. Sears, Phys. Rev. **185**, 200 (1969).
[5] G. C. Wick, Phys. Rev. **94**, 1228 (1954).
[6] A. S. Rinat, Phys. Rev. B **36**, 5171 (1987).
[7] J. Mayers, Phys. Rev. B **41**, 41 (1990).
[8] P. Martel, E. C. Svensson, A. D. B. Woods, V. F. Sears, and R. A. Cowley, J. Low Temp. Phys. **23**, 285 (1976).
[9] V. F. Sears, Can. J. Phys. **59**, 555 (1981).
[10] W. J. L. Buyers, V. F. Sears, P. A. Lonngi, and D. A. Lonngi, Phys. Rev. B **11**, 697 (1975).
[11] V. F. Sears, Phys. Rev. B **30**, 44 (1984).