Pseudoquasielastic component in the neutron scattering cross section

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Neutron scattering cross sections exhibit no quasielastic components when the scatterers move in one-dimensional Hamiltonian potentials. However, if the potential contains flattened regions, part of the inelastic spectrum appears as a pseudoquasielastic (PQE) component which could, in principle, be mistaken for a genuine quasielastic line. Using as a paradigm a power-law potential, we investigate the main features of this PQE component and discuss some of its properties (temperature, frequency dependence) that would help us to distinguish it from the quasielastic signal. Analytical results are given for the elastic and inelastic components.

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Neutron scattering is one of the most powerful tools used in the study of the geometry and dynamics of condensed systems[1–3]. As a function of the energy transfer $\hbar\omega$, the neutron cross section exhibits three regions: an elastic line, $\omega=0$, which informs about static properties; a quasielastic (QE) region (small ω), which contains the signatures of diffusional and other nonperiodic motions; and an inelastic part, related to vibrations. A careful study of the properties of the neutron scattering cross section in the case of scatterers moving in one-dimensional Hamiltonian potentials was presented ten years ago.[4] One of the main results of that study was that a QE component cannot emerge unless the potential recorded by the scatterer contains flat segments. This result was proved by calculating an upper bound for the QE component and showing that the scattered intensity strictly vanishes when the energy transfer $\hbar\omega$ goes to zero (although not for $\hbar\omega=0$, of course). However, the proof also indicated that motion in a potential that has a softer-than-quadratic extremum may generate a structure in the small energy transfer region that, although vanishing at $\omega=0$, could be mistaken for a bona fide QE component. We will call such a structure a pseudoquasielastic (PQE) peak. It would be of interest to study the genesis of the PQE peak as the potential is locally flattened. The problem has practical importance, because the PQE peak would appear superimposed to a genuine QE peak resulting, for instance, from the transference of intensity from the elastic line due to stochastic forces. It would be useful to have criteria to distinguish the signatures of both processes.

In this paper we clarify this issue, by calculating in detail the spectrum resulting from a scatterer of mass m moving in a power-law potential of the form $V(x)=A|x|^q$. This is an adequate paradigm for those situations in which a flattened region (which could also be a maximum) generates a PQE. We will show explicitly how the PQE component emerges from the *inelastic* spectrum as q is increased. Models containing flat regions were introduced in condensed matter by Kincaid and Eyring as early as 1937.[5] More recently, potentials that contain regions of lower-than-quadratic curvature have been used in the context of the soft potential model in glasses.[6]

Except for trivial factors, the cross section is given by the dynamical structure factor (DSF) $S(\kappa,\omega)$, where κ is the wave number transfer. Our starting point is the general expression for the DSF in terms of a superposition of energy integrals [see Eq. (2.11) in Ref. 4],

$$S(\kappa,\omega)=Z^{-1}\sum_{n=-\infty}^{\infty}\int_0^{\infty}dE\,e^{-\beta E}\tau(E)|F_n(\kappa,E)|^2 \times\delta[\omega-nW(E)], \qquad (1)$$

where $\tau(E)$ and $W(E)=2\pi\tau^{-1}(E)$ are, respectively, the oscillator period and its frequency, $\beta=(k_BT)^{-1}$ is the inverse temperature, Z is the partition function,

$$Z=\int_0^{\infty}dE\,e^{-\beta E}\tau(E), \qquad (2)$$

and the Fourier coefficients F_n are defined as

$$F_n(\kappa,E)=\tau^{-1}(E)\int_0^{\tau(E)}dt\,e^{-inW(E)t}e^{i\kappa x(E,t)}. \qquad (3)$$

Here $x(E,t)$ is the instantaneous scatterer location. The calculation of the period is standard.[7] For the chosen potential, we obtain

$$\tau(E)=PE^{1/q-1/2}, \qquad (4)$$

where

$$P=\frac{(8m)^{1/2}}{qA^{1/q}}B(1/q,1/2), \qquad (5)$$

$B(x,y)$ being the usual Beta function.[8] The partition function is

$$Z=\frac{(8\pi m)^{1/2}\Gamma(1/q)}{qA^{1/q}\beta^{1/2+1/q}}, \qquad (6)$$

where $\Gamma(x)$ is the Gamma function.[8]

The ω dependence of the DSF is determined by the structure of the frequency function $W(E)$ through the $E\rightarrow\omega$ mapping defined by the δ functions. The intensity $S_0(\kappa)$ of the elastic line is given by the $n=0$ term in Eq. (1). The integral projects the whole energy range onto the point $\omega=0$. To calculate the Fourier coefficient $F_0(E)$ it is convenient to write the solution to the equation of motion as

$$x(E,t)=X(E)G[4\Theta(q)t/\tau(E)], \tag{7}$$

where $X(E)=(E/A)^{1/q}$ is the location of the right-hand turning point and $\Theta(q)=q^{-1}B(1/q,1/2)$. A suitable change of variables and some algebra lead to

$$F_0(\kappa,E)=\frac{1}{\Theta(q)}\int_0^1 dv\frac{\cos[\kappa X(E)v]}{(1-v^q)^{1/2}}. \tag{8}$$

This is an oscillating function of E, whose oscillations are dampened with increasing E. The fluctuations are also softened with increasing q, i.e., with a hardening of the upper part of the potential. For large values of q these oscillations have a very long wavelength. As a consequence, $F_0(E)$ becomes almost energy independent over the whole relevant range of Eq. (1), leading to a very slow decrease of the elastic line with temperature. Indeed, in the limit case of the infinite square well, the size of the elastic line becomes temperature-independent [see Eq. (4.5) in Ref. 4].

Equation (8) can also be expanded in a power series that contains only even powers of $\kappa X(E)$. This series can be integrated term by term. We obtain

$$S_0(\kappa^q T/A)=\frac{\Gamma(1/2+1/q)}{[\Gamma(1/q)]^2}\sum_{i,j=0}^{\infty}R_iR_j\Gamma\left[\frac{1}{2}+\frac{2(i+j)+1}{q}\right]\times\left[\kappa\left(\frac{k_BT}{A}\right)^{1/q}\right]^{2(i+j)}. \tag{9}$$

Here

$$R_i=(-1)^i\Gamma[(2i+1)/q]\times\{\Gamma[(4i+2+q)/(2q)]\Gamma(2i+1)\}^{-1}.$$

The argument of S_0 indicates that, for a given q, the dependence on the physical variables appears only through the ratio $\kappa^q(T/A)$. In the low temperature region, $\kappa(k_BT/A)^{1/q}\ll 1$, we can retain only the first two terms, obtaining

$$S_0(\kappa^q T/A)\simeq 1-\frac{\Gamma(3/q)}{\Gamma(1/q)}\left[\frac{k_BT}{A}\right]^{2/q}\kappa^2. \tag{10}$$

For $q=2$, $S_0\simeq 1-(k_BT/2A)\kappa^2$, the simple harmonic oscillator result.[9] At $T=0$ all the intensity is in the elastic line; as the temperature is increased, the intensity is partially transferred to the rest of the spectrum. This can be seen in Fig. 1, where we observe the fast initial decay corresponding to high-q potentials (in all figures we take $A=1$). Since a high-q potential is very soft near its minimum, in the low-temperature domain the scatterer records a rapidly increasing region of configuration space as T is increased. Conversely, at high temperatures the size of the recorded region grows very slowly, leading to a weak decrease of the elastic intensity with temperature seen in the figure for large q. In the limit case $q\to\infty$, $S_0(\kappa)$ becomes temperature-independent for all $T>0$. The suitability of Eq. (10) as a low-temperature approximation is confirmed by the good agreement between the curves in the inset of Fig. 1.

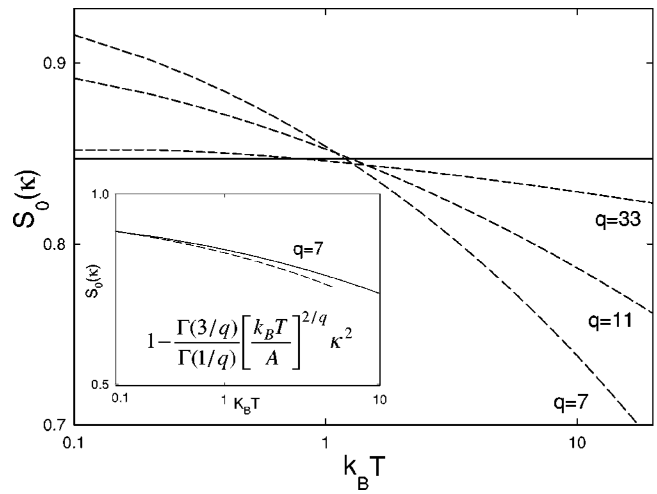

FIG. 1. Elastic line intensity as function of the temperature for $\kappa=0.7$ and the values of q indicated in the figure. The solid line is the result for the infinite square well. Inset: the small T approximation (dashed line). The units are arbitrary.

The dependence of $S_0(\kappa)$ with κ can be analyzed in the same manner, since κ and T always appear combined as the product κ^qT. In particular, $S_0(\kappa)$ is a monotonically decreasing function of κ.

Because of the delta functions in Eq. (1), only those energy domains for which the oscillation frequency is small contribute to the DSF in the small ω region. In the case of a power-law potential, the relevant domain corresponds to energies close to the potential minimum. Higher intensities result when the $E\to\omega$ mapping projects a large energy domain onto a very small segment along the ω axis. From Eq. (4), we see that only a narrow energy domain contributes to the $\omega\to 0$ region, but that its size grows for higher values of q. By using the delta function in Eq. (1), it is easy to see that, for $q>2$, the DSF in the $\omega>0$ region is given by

$$S_{in}(\kappa,\omega)=\frac{2qP}{|q-2|Z}\sum_{n=1}^{\infty}\left(\frac{P}{2\pi n}\right)^{\frac{q+2}{q-2}}\times\omega^{4/(q-2)}e^{-\beta E_n^*}|F_n(E_n^*)|^2, \tag{11}$$

where $E_n^*=(P\omega/2\pi n)^{2q/(q-2)}$. Through suitable variable changes, the Fourier coefficients can be put into the form

$$F_n(E)=\frac{1}{2\Theta(q)}\int_{-1}^{1}\frac{dv\,e^{i\kappa vX(E)}}{(1-|v|^q)^{1/2}}\times\cos\left[\frac{n\pi}{2\Theta(q)}\int_{-1}^{v}\frac{d\xi}{(1-|\xi|^q)^{1/2}}\right]. \tag{12}$$

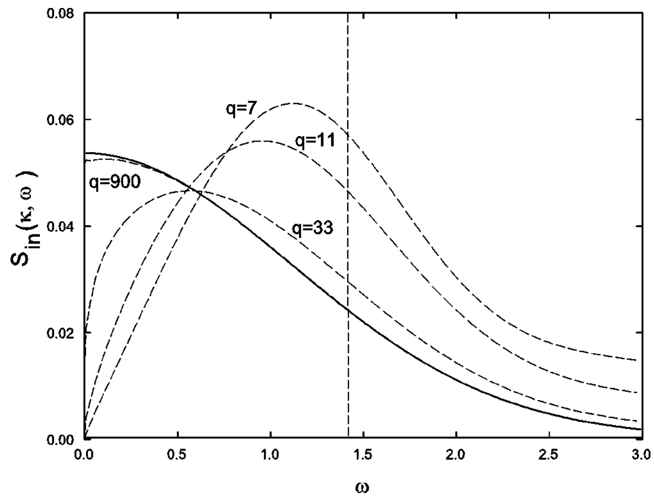

FIG. 2. Inelastic intensity as a function of frequency for $\beta=2$ and $\kappa=0.7$, and the values of q indicated in the figure. The solid line corresponds to the infinite square well. The dotted vertical line corresponds to the first resonance for the harmonic oscillator ($q=2$).

These formulas are exact. The v integral may be simplified if we note that the coefficient of the exponential is even in v for n even and odd for n odd. It can then be shown that $F_n(E_n^*)$ becomes ω-independent as $\omega\rightarrow 0$. In this limit, the DSF is therefore a sum of terms of the form

$$S_{in}^{(n)}(\kappa,\omega\rightarrow 0)\sim\omega^{4/(q-2)}\exp\left[-\frac{1}{k_BT}\left(\frac{P\omega}{2\pi n}\right)^{2q/(q-2)}\right]. \tag{13}$$

In the range of our interest ($q>2$), $S_{in}(\kappa,\omega\rightarrow 0)\sim\omega^{4/(q-2)}$. This indicates that the depression of the spectrum in the small ω region weakens as q grows, i.e., as the potential bottom flattens, a PQE structure arises. As seen from Eq. (4.5) in Ref. 4, in the limit case of a square-well potential with infinitely high walls, $S_{in}(\kappa,\omega)$ becomes a sum of Gaussians. The exact form of $S_{in}(\kappa,\omega)$ is depicted in Fig. 2, where we see that, with increasing q, more of the inelastic intensity is transferred to lower frequencies. For high values of q, the inelastic peak converges towards the result for the infinite square well, except for a small neighborhood of $\omega=0$, where it must agree with Eq. (13). Our numerical evaluations (not shown) also indicate that Eq. (13) is an excellent approximation. The first resonance for the harmonic oscillator, represented in Fig. 2, can be used as a standard to ascertain the location of the PQE component. For instance, if this resonance corresponds to $\omega_0=10^{12}$ Hz, then the PQE component for $q=33$ and the same value of A lies in the interval 10^{10}–10^{12} Hz.

A different aspect of the problem is illustrated in Fig. 3, where we observe how the intensity distribution as a function of κ evolves with increasing q towards that corresponding to the infinite square well. By increasing q, the stronger confinement leads to the growth of the main peak and to its shift towards shorter wave numbers.

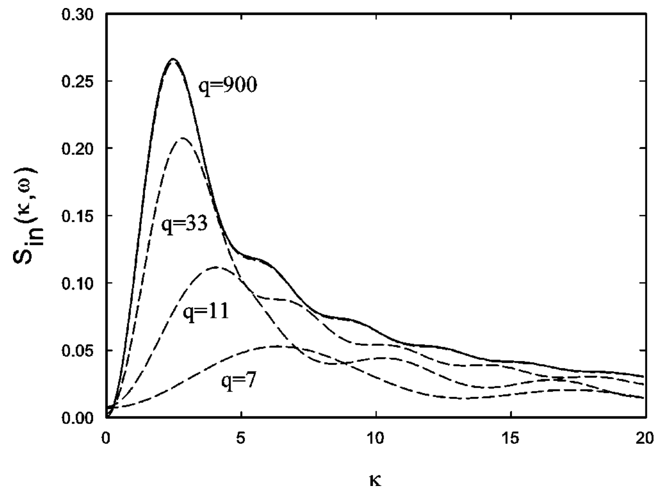

FIG. 3. Wave number dependence of the inelastic intensity for $\beta=2$ and $\omega=0.1$, and the values of q indicated in the figure. The solid line corresponds to the infinite square well.

How do genuine QE peaks arise? For pure diffusion, the motion of the scatterer is not confined and the elastic line disappears completely. The DSF is a Lorentzian whose half-width is $2\kappa^2 D$, the temperature dependence entering through the diffusion coefficient D. The classical case of a QE component arising from restricted motion is the harmonic oscillator subject to frictional forces. Because of the confinement there is a surviving elastic line; the QE component is formed by a superposition of Lorentzians whose widths are proportional to the friction coefficient. The decomposition of the QE signal into Lorentzians is also possible for more general potentials—at least for those having harmonic bottoms.[10] As noted before, the total QE intensity can be obtained as the difference between the elastic intensity and the Debye-Waller factor.[4,6]

Stochastic forces cause intensity to be transferred from the elastic line into the QE region. Flattening potentials cause a shift of the inelastic spectrum towards the frequency origin. We summarize the main differences between the resulting structures.

(a) The QE tends to a positive constant as $\omega\rightarrow 0$; in this limit the PQE tends to zero as a power law.

(b) The QE has a long (Lorentzian) tail for large ω. The PQE has no such a long tail, its nth component being bounded by the fast-decaying function $\exp[-\beta(\omega/2\pi n)^{2q/(q-2)}]$.

(c) The n-th Lorentzian in the QE depends on temperature as $T^{2n}\exp[-(\kappa/\omega_0)^2T]$. The nth component in the PQE is proportional to $T^{-(1/2+1/q)}\times\exp(a_n/T)$, where a_n is a function of q and T that can be obtained from Eq. (11).

Next we show that the magnitude of the predicted PQE component is commensurate with that resulting from well-known scattering potentials. This is conveniently assessed by considering two different benchmarks.

(a) The Hamiltonian infinite square well. Since the scattered intensity for large q resembles that corresponding to an infinite square well, the magnitude of the square well inelastic scattering is a suitable measure of the size of the PQE effect. From Eq. (4.5) in Ref. 4, we see that the integrated inelastic intensity for a square well of width d is $i(\kappa)=1-[(2/\kappa d)\sin(\kappa d/2)]^2$. For typical values of d, say, 2–3 Å, $i(\kappa)$ will oscillate between 0 and unity as κ is varied in the experimentally accessible range.

(b) The harmonic oscillator subject to frictional forces. This problem was studied in detail in Ref. 10. There it was shown [Eq. (59)] that the total QE intensity is $i_{QE}=\exp(-R)[I_0(R)-1]$, where $R=T\kappa^2/2A$, and I_0 is a modified Bessel function. The i_{QE} maximum is at $R\approx 3$ and corresponds to about 20% of the total scattered intensity. From Figs. 1 and 2, we see that 20% is also a typical value of the fraction of intensity scattered in the PQE region for the potentials studied in this paper.

Let us conclude by noting the PQE is robust in the sense that it should be observable even if the system contains a heterogeneous distribution of scatterer parameters $\{A,q\}$.

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