Effects of phonon transfer on near-thermal neutron fission cross sections

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For low energy neutron-induced fission in the thermal and epithermal range, the momentum transferred in forming the compound nucleus is small enough that the compound nucleus may remain bound in the lattice until it decays. Phonons, therefore, may be emitted or absorbed in the neutron absorption process changing the energy of the compound nucleus from that which would be derived simply from measuring the incoming neutron energy. The probability and influence of phonon transfer to and from the lattice is calculated at energies below 1 eV and is shown to have a small but significant effect on the observed cross section. The magnitude of the effect is temperature dependent and ranges in size from a few tenths of 1% for ^{235}U fission at thermal to 5% for ^{239}Pu fission at the 0.3 eV resonance. Some of the effect can be accounted for by applying the usual Doppler broadening approximation in the thermal range.

[NUCLEAR REACTIONS FISSION Capture, Doppler effect, phonon exchange; temperature dependence.]

BASIC FORMALISM

From the earliest days of neutron physics it was recognized that the thermal motion associated with the physical conditions of a sample would influence the neutron cross section for various reaction processes.[1,2] Lamb[3] in 1939 showed that for a nucleus bound in a lattice, phonon absorption and emission would influence the compound nucleus energy and therefore the nuclear cross section. He laid the basis for performing such evaluations and applied it to the broadening of neutron resonances in the weak binding approximation now commonly referred to as the gas approximation of Doppler broadening. Subsequently several other authors have developed these concepts further for broadening of neutron resonances.[4,5]

The Lamb theory was used also by Mössbauer[6] to explain the recoilless emission and absorption of γ rays. Visscher[7] carried out calculations emphasizing the possibility of studying crystal lattice structure via the Mössbauer effect. In the course of his development he demonstrates a method for expansion of the reaction probability in terms of discrete numbers of phonons emitted or absorbed, which is followed closely here.

The effects of the physical properties of the sample on resonance shape, therefore, have been studied rather thoroughly and significant effects are detected experimentally. In fact almost all neutron resonance analysis takes Doppler broadening into account assuming a "gas approximation." Jackson and Lynn[5] have carried out calculations showing more detailed effects of crystalline structure and have verified their existence at the few percent level by experiment. However, in spite of the fact that on a fractional basis energy exchange becomes larger as the neutron energy decreases, to the knowledge of the authors, no calculations have been performed to evaluate such effects in the thermal and epithermal range.

In the course of studies at NBS to improve the accuracy of thermal cross section standards and to normalize keV cross section measurements on ^{235}U, a study of the influence of phonon transfer on fission and other reaction cross sections has been carried out. The intent of this paper is to develop a formalism for calculating the influence of phonon exchange effects in fission and other reaction cross sections at low energies, and to calculate the magnitude of the effects for a few interesting examples.

DERIVATION

We follow closely the derivation of Lamb for capture and then generalize it to the fission case. We wish to find the probability $W(\{\beta_s\};\{\alpha_s\})$ for the absorption of a neutron of momentum $\vec{p}$ by a definite lattice atom L of type A to form a nucleus of type B with the emission of a γ ray of wave vector $\vec{k}$ when the crystal undergoes a transition from a state $\{\alpha_s\}$ to a state $\{\beta_s\}$. The set of numbers $\{\alpha_s\}$ gives the number of phonons in each of the s modes of oscillation of the crystal lattice before the neutron is absorbed and $\{\beta_s\}$ is the corresponding notation for the lattice state after emission of a capture γ ray.

The final state is reached through a compound nucleus state C during which the lattice is in the state $\{n_s\}$. According to Lamb the required probability W near a nuclear resonance can be written

$$W(\{\beta_s\};\{\alpha_s\}) = \left|\sum_{n_s} \frac{(B\beta_s\vec{k}|H''|Cn_s)(Cn_s|H'|A\,\alpha_s\vec{p})}{E_0 - E + E(n_s) - E(\alpha_s) + \frac{1}{2}i\Gamma(n_s,\alpha_s)}\right|^2, \quad (1)$$

where E is the laboratory kinetic energy of the neutron, E_0 is the energy of the resonance if the target is rigidly bound or infinitely heavy, and $\Gamma(n_s,\alpha_s)$ is the full width at half maximum of the resonance. The quantity $E(\alpha_s)$ can be written

$$E(\alpha_s) = \sum_s \hbar\omega_s(\alpha_s + \tfrac{1}{2}), \quad (2)$$

and a similar definition applies for $E(n_s)$. These values are generally much larger than E, but their difference is generally smaller than E, or, for low energies, more nearly comparable.

The matrix element containing H' can be factored into a nuclear part for compound nucleus formation and a part describing the transfer of momentum to a particular atom with excitation of the lattice via this interaction from the state $\{\alpha_s\}$ to the state $\{n_s\}$. The matrix element containing H'' can be factored into a nuclear part for the decay of the compound nucleus by γ-ray emission and a part describing the transition of the lattice via momentum transfer from the state $\{n_s\}$ to a state $\{\beta_s\}$. The sum ranges over all states $\{n_s\}$ which can be reached from the array of possible initial states $\{\alpha_s\}$.

In the derivation of Eq. (1) it is assumed that[3,7] the state of the crystal associated with the compound nucleus, $\{n_s\}$, remains unchanged during the compound nucleus lifetime. It is therefore valid only when the relaxation time of the crystal is long compared with the lifetime $\hbar/\Gamma$ of the compound nucleus. The relaxation lifetime of a crystal lattice is typically about 10^{-12} sec. By comparison the lower limit to the total neutron resonance widths is ~20 meV for the heaviest nuclei which do not undergo slow neutron fission. The corresponding lifetime is 3×10^{-13} sec so that lifetime requirements are well satisfied. When fission also is present, the lifetime is reduced typically by another order of magnitude.

In practice the lattice is not in a definite state $\{\alpha_s\}$ but is in thermal equilibrium so that a wide spectrum of initial lattice states are possible. The probability of a particular state will be denoted by $g(\{\alpha_s\})$ and an average must be taken over these initial states. In addition, if we are interested in the process for decay by γ-ray emission to a given nuclear level i, in almost any practical experiment the emitted γ-ray energy is far larger than the lattice excitation and the energy resolution now experimentally possible does not permit the detection of γ rays leaving the lattice in some particular state $\{\beta_s\}$. We must then sum over the final lattice states $\{\beta_s\}$ to arrive at a measurable quantity. Therefore we take the average over the initial states and the sum over the final states to find the probability $W_i(E)$ for decay to the state i to be

$$W_i(E) = \sum_{\beta_{si}} \sum_{\alpha_s} g(\{\alpha_s\}) W(\{\beta_s\};\{\alpha_s\}). \quad (3)$$

As mentioned earlier the matrix elements of Eq. (1) can be factored and, upon performing the sum over β_{si} of Eq. (3), $W_i(E)$ can be written

$$\frac{W_i(E)}{(|M_{\gamma i}|^2)} \equiv W'(E) = \sum_{\alpha_s} g(\alpha_s) \sum_{n_s} \frac{|(n_s|\exp(i\vec{p}\cdot\vec{\chi}_L/\hbar)|\alpha_s)|^2 |M_c|^2}{[E - E_0 - \sum_s (n_s - \alpha_s)\hbar\omega_s]^2 + \frac{1}{4}\Gamma^2(n_s,\alpha_s)}, \quad (4)$$

where $M_{\gamma i}$ is the nuclear matrix element for primary γ-ray emission to the state i and M_c is the matrix element for compound nucleus formation. The matrix element within the sum represents the excitation from initial state $\{\alpha_s\}$ to a compound nucleus state $\{n_s\}$ through the absorption of momentum $\vec{p}$ by the Lth atom at position χ_L in the lattice. We have substituted with Eq. (2) to obtain the form of the denominator.

The quantity $|M_c|^2$ can be shown to be equal to the neutron width Γ_n and the quantity $|M_{\gamma i}|^2$ to the radiation width to the ith level $\Gamma_{\gamma i}$. The cross section for neutron capture followed by γ-ray decay to the state i defined by $\sigma_{\gamma i}$ can be shown[7] to be related to $W'(E)$ by

$$\sigma_{\gamma i} = 4\pi\lambda\!\!\!^{-2} g \Gamma_{\gamma i} W'(E), \quad (5)$$

where g is the statistical factor and $\lambda\!\!\!^{-}$ is the reciprocal wave number. Upon finding a means to perform the sums of Eq. (4), the influence of phonon transfer on a partial capture cross section can be calculated. Note that nothing within the sums of Eq. (4) depends on the final state. The total capture cross section therefore can be obtained by further summing, over all possible states to which γ rays initially can be emitted:

$$\sigma_\gamma = \sum_i \sigma_{\gamma i} = 4\pi\lambda\!\!\!^{-2} g \sum_i \Gamma_{\gamma i} W'(E) = 4\pi\lambda\!\!\!^{-2} g \Gamma_\gamma W'(E). \quad (6)$$

The generalization to include fission follows the same arguments used above to proceed from compound nucleus decay to a particular nuclear state i and to the capture cross section expressed by Eqs. (4) and (6). In fission the number of final states is very large when we consider the internal excitation of the two fragments, the neutron emission spectrum, and lattice excitations. In the same way as described before with Eq. (3), we perform a weighted average over the possible initial lattice states $\{\alpha_s\}$ and sum over the possible final states $\{\beta_s\}$ since the influence of the various final lattice states cannot be detected by measuring the high energy products from fission. A nuclear matrix element for a given final state configuration i can also be written for fission, called M_{fi} and substituted for $M_{\gamma i}$ in Eq. (4). To obtain the fission cross section for production of a particular mass pair, we must further sum over all configurations possible for the pair. To obtain the total fission cross section we finally sum over all possible mass pairs. By following this sequence we demonstrate an evaluation of $|M_{fi}|^2 \rightarrow \Gamma_f$ and by analog to Eq. (6) find

$$\sigma_{nf} = 4\pi\lambda\!\!^{-2}\Gamma_f W'(E)\,. \tag{7}$$

Returning to Eq. (4) for $W'(E)$ we see that the sum is made difficult to handle by three factors; the sum over (n_s, α_s), the dependence of Γ on α_s and n_s, and the dependence of $|M_c|^2 = \Gamma_n$ on α_s and n_s. The width Γ_n is the only nuclear parameter with significant dependence on neutron energy and phonon-transfer effects. For heavy nuclei in the energy range below a few eV, it is, however, very much smaller than Γ_γ or Γ_f so that the energy dependence of Γ_n does not significantly affect Γ. For the purpose of this development Γ is taken to be constant.

In doing so we follow Lamb[3] who pointed out the dependence of Γ on phonon transfer effects through the neutron width Γ_n. For the case he was interested in, however, of resonances of several eV energy or higher, the effect of phonon transfer was negligible not because the neutron width was so small, but because the energy transferred to or from the lattice is so small compared to the neutron's energy.

The energy region of concern for us is below 1 eV where the effects of phonon transfer on Γ in the denominator are negligible but the effects on Γ_n in the numerator are not. We perform the calculation under two conditions. In the first we neglect the phonon transfer on both Γ_n and Γ which we will call "Lamb's approximation." In the second, we neglect the dependence on Γ but take into account the dependence on Γ_n which we call "Γ_n Dependence."

LAMB'S APPROXIMATION

The dependence on $(n_s - \alpha_s)$ significantly complicates the summing process of Eq. (4). Lamb handles this by grouping together terms with the same value for $\sum_s (n_s - \alpha_s)\hbar W_s$ and adding them using a δ function method. He also demonstrates that for the matrix element in the numerator, all terms are zero except those corresponding to processes whereby the occupation number of any particular phonon state is changed at most by ±1. Many phonons may be transferred in a particular reaction but only one is possible to or from a particular phonon state. The sum over n_s is then taken and finally the sum over $g(\alpha_s)$ using the average value of α_s in each of the s cases as derived from Bose statistics at thermal equilibrium,

$$\alpha = [\exp(\hbar\omega/kT) - 1]^{-1}\,. \tag{8}$$

The result of performing the sums of Eq. (4) is then found to be

$$W'(E) = 2/\Gamma \,\mathrm{Re} \int_0^\infty d\mu \exp[i\mu(E - E_0 + \tfrac{1}{2} i\Gamma + g(\mu)]\,, \tag{9}$$

where $g(\mu)$ is given by

$$g(\mu) = \frac{m}{M}\frac{E_n}{3N}\int_0^{\omega_{\max}} \frac{N(\omega)}{\omega} d\omega[\alpha e^{i\mu\omega} + (\alpha+1)e^{-i\mu\omega} - 2\alpha - 1]\,, \tag{10}$$

where in Eq. (10) the sum has been replaced by an integral and $3N$ is the number of degrees of freedom of a crystal containing N atoms.

The term in $g(\mu)$ containing the quantity $e^{i\mu\omega}$ represents reactions in which phonons are gained from the lattice, the term with $e^{-i\mu\omega}$ represents reactions where phonons are lost to the lattice and the remaining terms are for no phonon exchange. By substituting a phonon frequency spectrum $N(\omega)$, $g(\mu)$ can be calculated and then using (9) a cross section modified by phonon-transfer effects can be derived.

The Debye spectrum

$$N(\omega) = 9N\omega^2/\theta^3 \tag{11}$$

shows a quadratic form up to a cutoff frequency $\omega_{\max}$ equal to the Debye temperature θ. (In the remainder of this report when referring to energies $\hbar\omega$, $k\theta$, and kT, Planck's constant and the Boltzmann constant will be dropped.) The normalization is such that $\int_0^{\omega_{\max}} N(\omega)d\omega = 3N$. It is not possible to obtain a simple analytic expression for the cross section using this spectrum, which is generally useful at all temperatures. However, in the $T \rightarrow 0$ limit, a simple analytic solution is possible. For higher temperatures we use the

Einstein approximation which assumes that all phonons have the same energy ϵ. This δ function approximation of the frequency spectrum may be a better approximation of the actual phonon distribution than the Debye approximation in many cases. Both of these solutions will be derived in the next section of the report.

$T=0$ LIMIT

By reference to Eq. (8) we see that, when $T=0$, that $\alpha=0$ so that the integrand in $g(\mu)$ is greatly simplified to

$$e^{-i\mu\omega}-1\,. \tag{12}$$

Note that only the term permitting phonon loss to the lattice and a no-exchange term remain. Obviously at $T=0$, since there are no phonons in the crystal, no phonon can be transferred from the lattice.

We next break $g(\mu)$ into two parts $g(\mu)=g_l(\mu)+g_0(\mu)$ where the subscripts l and 0 refer to phonon loss and no exchange, respectively, and substitute the Debye spectrum, Eq. (11), and integrate to obtain the expressions

$$g_l(\mu)=\frac{3m}{M}\frac{E_n}{\theta^3}\left[\frac{\theta e^{-i\mu\theta}}{-i\mu}+\frac{1}{\mu^2}\left(e^{-i\mu\theta}-1\right)\right], \tag{13a}$$

$$g_0(\mu)=\frac{3}{2}\,\frac{m}{M}\,\frac{E_n}{\theta}\,. \tag{13b}$$

In order to carry out the integral required in Eq. (9), we expand the term in Eq. (9) $e^{g(\mu)}$ as below:

$$e^{g(\mu)}=e^{g_0}e^{g_l(\mu)}$$
$$=e^{g_0}\left(1+g_l(\mu)+\frac{g_l(\mu)^2}{2}+\cdots\right). \tag{14}$$

Following Visscher[7] we identify the first term in the expansion as the no-phonon-loss term, the second term $g_l(\mu)$ as the one-phonon-loss term, the third term as the term for two phonons lost, etc. If the neutron energy is small (<0.5 eV for heavy nuclei), it can be shown that the above expansion converges rapidly. On the other hand, in the eV range the convergence is too slow to make such an expansion useful. Lamb derived the "gas" approximation for Doppler broadening of resonances by an approximate solution to Eq. (9). The primary departure of this work from Lamb's and other work is to use the expansion of Eq. (14) to derive solutions useful for describing phonon effects on nuclear reaction cross sections at low neutron energies.

The expansion gives rise to a series of terms $W_0'(E)=W_{0,0}'(E)+W_{0,1}'(E)+W_{0,2}'(E)+\cdots$, where subscripts 0, 1, and 2 refer to the numbers of phonons lost in the interaction and the other subscript to the $T=0$ approximation. The calculation for $W_{0,2}'$ cannot be expressed in a simple analytic form. However, since $W_{0,1}'$ is much smaller than $W_{0,0}'$ at low energies, the correction for neglect of $W_{0,2}'$ is negligible at low energies. Upon carrying out the integration for W' we find

$$W_{0,0}'(E)=e^{g_0}\frac{1}{(E-E_0)^2+\frac{1}{4}\Gamma^2}, \tag{15}$$

$$W_{0,1}'(E)=\frac{6mE_ne^{g_0}}{M\theta^3\Gamma}\times\left[\frac{1}{4}\ln\frac{(E-E_0-\theta)^2+\frac{1}{4}\Gamma^2}{(E-E_0)^2+\frac{1}{4}\Gamma^2}+(E-E_0)\tan^{-1}\times\frac{\frac{1}{2}\Gamma\theta}{[E-E_0)^2-\theta(E-E_0)+\frac{1}{4}\Gamma^2]}\right]. \tag{16}$$

The second term in $W_{0,1}'$ is valid for $(E-E_0)^2+\frac{1}{4}\Gamma^2>\theta(E-E_0)$. Appropriate values of $\pm\pi$ must be added otherwise to assure continuity of the term across the angles $\pm\frac{1}{2}\pi$.

Although it is not obvious, the two terms in $W_{0,1}'(E)$ almost cancel. For distant resonances we find

$$W_{0,1}'(E)=-g_0e^{g_0}[(E-E_0)^2+\tfrac{1}{4}\Gamma^2]^{-1}.$$

The ratio of phonon transfer to the rigidly bound case can be calculated from the product $W'(E)[(E-E_0)^2+\frac{1}{4}\Gamma^2]$. In Fig. 1 this quantity for ^{235}U calculated with $T=0$ using Eqs. (15) and (16) is shown by the dashed line. The Debye temperature is taken to be $\theta=16$ meV and resonances for ^{235}U resonances are assumed at −0.916, −0.020, and +0.287 eV. The calculation is carried out by adding the effects

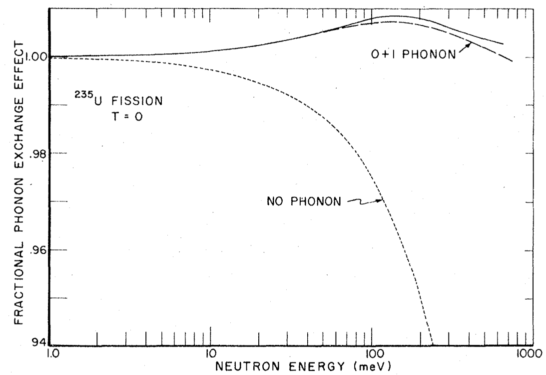

FIG. 1. The quantity $W'(E)[(E-E_0)^2+\frac{1}{4}\Gamma^2]$, which is the ratio of the ^{235}U fission cross section with phonon exchange to the rigidly bound case, is shown as a function of neutron energy for temperature $T=0$ K. The dotted line shows the no-phonon-exchange portion. The dashed line shows the cumulative total of zero and one phonon exchange and the solid line the total of all phonon-transfer processes.

of single levels without interference. The error of neglecting multilevel interference is negligible when the *ratio* is calculated for cross sections with and without phonon-transfer effects. The dotted line shows the no-phonon-transfer portion calculated from e^{g_0}. The difference between the dashed and dotted curves gives the effect of one phonon transfer. The solid line shows the results of a computer calculation which uses an arbitrary number of terms in the expansion of Eq. (14) to obtain the total phonon-transfer effect. It appears that such a calculation is possible only for $T=0$ when a Debye spectrum is used. The difference between the solid and dashed lines gives the magnitude of the transfer of two or more phonons.

Several implications of Fig. 1 are of interest. First, the effect of taking into account phonon transfer is not entirely negligible compared with the presently assumed accuracy of the cross section, which lies in the 0.5% range.[8] The behavior of the effect will be of different sign and magnitude for other nuclei depending on the role of resonance position and strengths. Note that there is no single $T=0$ cross section for ^{235}U. The shape of the cross section depends on the Debye temperature or, more exactly, on the frequency spectrum associated with the particular chemical form of the ^{235}U. There is then in general a chemical dependence of low energy fission and other reaction cross sections, which persists even at $T=0$. The dependence is not associated with the zero-point vibration of the atoms at $T=0$, but rather by the spectrum of higher lying phonon states which can be excited by absorption of energy from the neutrons.

Additional physical insight is provided by the carry over of Lipkin's sum rule[9] for Mössbauer photon interactions to the neutron case. The sum rule restated for neutrons says that the average energy transferred by the neutron is the same regardless of chemical form or sample temperature. As the neutron energy goes to zero, only in rare events is any energy transferred (i.e., phonons excited). However, in those events the energy transferred is much greater than the average energy. This can be seen in Fig. 1 which shows that for ^{235}U at $T=0$, 99.4% of the neutron interactions at room thermal energy are recoil-free. The remaining 0.6% carry the energy which balances Lipkin's sum rule. The phonon exchange results in a loss in energy available to the compound nucleus. The reaction probability is effectively higher than otherwise owing to the dominance at low energies of the bound levels in ^{235}U. The cross section with phonon exchange is therefore higher by 0.3% than it otherwise would be.

HIGHER TEMPERATURES

While the $T=0$ approximation simplifies calculations and provides estimates of the magnitude of phonon exchange effects on low energy cross sections, the effects at higher temperatures are of greater practical interest. We show next how these may be calculated in analytic form using suitable approximations.

Returning now to Eq. (10) we seek a solution to the integral which will permit the integration of Eq. (9). A rather close approximation to the Debye spectrum is given by the expression

$$N(\omega)=\frac{3N}{T^2L}\,\omega(e^{\omega/T}-1)\,, \tag{17}$$

where L, a normalization constant necessary to satisfy the condition $\int_0^\theta N(\omega)d\omega=3N$, is given by

$$L=[e^{\theta/T}(\theta/T-1)-\tfrac{1}{2}(\theta/T)^2+1]\,. \tag{18}$$

This expression can be used to obtain useful results for zero- and one-phonon exchange for temperatures in the range $0<\theta/T<1$. The approximation improves as the temperature increases. However, since more than one phonon exchange is not readily handled, this approximation is of limited usefulness.

The Debye spectrum does not appear to approximate closely the measured or theoretically calculated frequency spectra for the actinides anyway. Experimental and theoretical studies[5,10,11] on UO_2, UC, and uranium metal show primarily two discrete lines in the spectra with a small tail extending down to zero energy. Jackson and Lynn[5] have shown that a single line or a pair of lines can be used to calculate adequately Doppler broadening for the resonance region. In this paper we approximate the spectrum with a δ function positioned at the energy which appears to be appropriate from other theoretical and experimental work. Calculations by Lajeunesse, Moore, and Yeater[11] for UC indicate a pair of peaks centered at 17 meV and Young[10] calculates for UO_2 a pair of peaks centered at about 14 meV. Jackson and Lynn[5] fitted the shape of the 6.67 eV resonance in ^{238}U metal with a single line at 11 meV. In subsequent calculations we use δ functions positioned at these three energies.

The function $N(\omega)=3N\delta(\omega-\epsilon)$ is substituted into Eq. (10) and the integral performed to give

$$g(\mu)=\frac{mE}{M\epsilon}[\alpha\, e^{i\mu\epsilon}+(\alpha+1)e^{-i\mu\epsilon}-2\alpha-1]\,, \tag{19}$$

where $\alpha=(e^{\epsilon/T}-1)^{-1}$. Substituting Eq. (19) into Eq. (9)

$$W'(E) = \frac{2}{\Gamma} \exp[-b(2\alpha+1)] \times \mathrm{Re} \int_0^\infty d\mu \, \exp i\mu[(E-E_0)+\tfrac{1}{2}i\Gamma] \times \{\exp(b\alpha e^{i\mu\epsilon}) \exp[b(\alpha+1)e^{-i\mu\epsilon}]\}, \quad (20)$$

where $b = mE/M\epsilon$. Expanding the last two exponentials of Eq. (20), multiplying and combining terms with the same values of $e^{-in\mu\epsilon}$ or $e^{+in\mu\epsilon}$ gives the product

$$\exp(b\alpha e^{i\mu\epsilon})\exp[b(\alpha+1)e^{-i\mu\epsilon}] = \sum_{n=0}^{\infty} e^{in\mu\epsilon} \sum_{j=0}^{\infty} \frac{[b(\alpha+1)]^j (b\alpha)^{j+n}}{j!\,(j+n)!} + \sum_{n=1}^{\infty} e^{-in\mu\epsilon} \sum_{j=0}^{\infty} \frac{[b(\alpha+1)]^{j+n}(b\alpha)^j}{j!\,(j+n)!}. \quad (21)$$

As in the low temperature case, the expansion has been carried out in terms of the number, $\pm n$, of phonons transferred. Equation (21) can be rewritten in terms of the Bessel function of the first kind of imaginary argument in the form

$$\exp(b\alpha e^{i\mu\epsilon})\exp[b(\alpha+1)e^{i\mu\epsilon}] = \sum_{n=-\infty}^{+\infty} e^{in\mu\epsilon}(\alpha/\alpha+1)^{n/2} I_{|n|}\{2b[(\alpha+1)\alpha]^{1/2}\}. \quad (22)$$

Substituting Eq. (22) into Eq. (20) gives

$$W'(E) = e^{-b(2\alpha+1)} \sum_{n=-\infty}^{+\infty} \frac{[\alpha(\alpha+1)]^{n/2} I_{|n|}\{2b[\alpha(\alpha+1)]^{1/2}\}}{(E-E_0+n\epsilon)^2+\frac{1}{4}\Gamma^2}. \quad (23)$$

$W'(E)$ is therefore given as an expression in terms of the number of phonons exchanged. This expression can be shown to correspond exactly to that of Eq. (14) in Jackson and Lynn's[5] paper with the exception of a typographical error in their δ function which should read $\delta(E_t - nh\nu_E)$ in their notation.

Equation (23) therefore is not original, but it does verify their result using a somewhat different and perhaps more transparent approach. The form of Eq. (23) also makes it clear that the phonon-transfer effects can be calculated as a sum of Breit-Wigner shapes displaced from one another by the increments of energy, $n\epsilon$, and weighted with the proper coefficient. The $n=0$ term corresponds to no phonon exchange. The $n=-1$ and $+1$ terms correspond, respectively, to one phonon lost to or gained from the lattice. In nearly all cases the $n<0$ terms will dominate. The envelope of the coefficients in the numerator corresponds at higher energies to the shape of the commonly used Doppler-broadening function.

We wish now to compare the fission cross section with phonon exchange included with the case

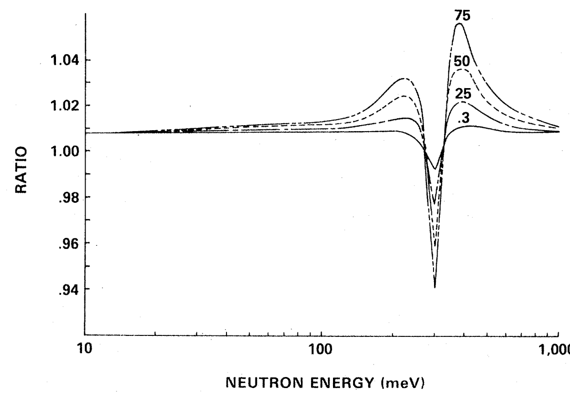

FIG. 2. The fractional phonon exchange effect for the fission cross section of ^{239}Pu is shown multiplied by the unbroadened Breit-Wigner denominator. For the Einstein model an $\epsilon = 14$ meV is assumed. The four curves show the effect as a function of temperature in meV. No influence of phonon transfer on Γ_n is included.

where the energy dependence is carried only in the usual Breit-Wigner denominator. Therefore, a convenient measure of the size of the phonon-exchange effect is the calculation of the quantity $W'(E)[(E-E_0)^2+\frac{1}{4}\Gamma^2]$.

This quantity is shown in Figs. 2 and 3 for ^{239}Pu and ^{235}U, respectively, as a function of neutron energy for several sample temperatures with T in meV. For ^{239}Pu only the resonance at 0.3 eV was included in the analysis. In both cases a value for $\epsilon = 14$ meV was used. The curves are essentially independent of the value of ϵ when it is varied by ±50% around the $\epsilon = 14$ value. This suggests an independence of chemical form in this energy region and at these temperatures at least as regards the influence of phonon transfer. The temperature dependence is significant relative to desired accuracy in the thermal range apparently indicating the need for care in handling thermal

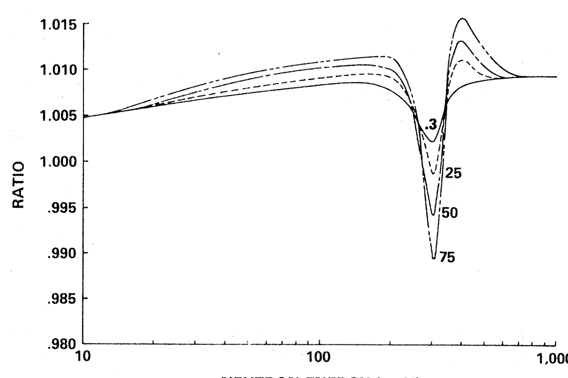

FIG. 3. The fractional phonon exchange effect for the fission cross section of ^{235}U is shown as calculated for Fig. 2.

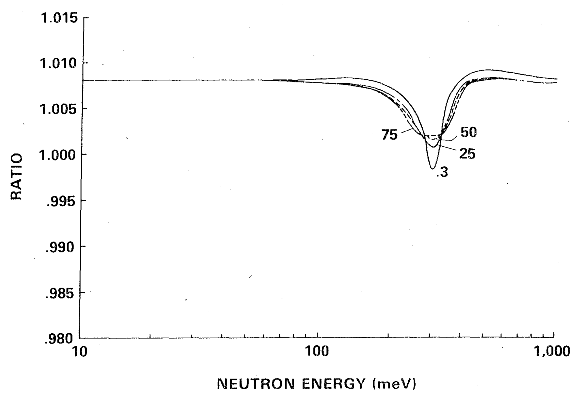

FIG. 4. The fractional phonon exchange effect for the fission cross section of ^{239}Pu is shown multiplied by the Doppler broadened and recoil shifted Breit-Wigner denominator. See Fig. 2 for details.

broadening regardless of the calculational technique applied.

The conventional Doppler broadening technique uses an effective temperature T' in accordance with Lamb's work[3] where we have taken the Debye temperature to be 13.75 meV. If we divide our $W'(E)$ by this quantity instead of the unbroadened Breit-Wigner curve as before, we get significantly different results. These are presented in Figs. 4 and 5 for ^{239}Pu and ^{235}U. We see that for ^{235}U the result becomes essentially temperature independent although residual influences of phonon transfer remain including the 0.5% displacement at thermal energy. For ^{239}Pu some small temperature dependence remains. In summary, the calculation demonstrates a temperature dependence owing to influences on the Breit-Wigner denominator which is not negligible at thermal energy. However, the conventional Doppler broadening technique provides a correction which follows closely the calculation here.

The Westcott g factor[12] which is used to relate spectrum-averaged fission rate to rates with monoenergetic neutrons and to derive temperature dependent reaction cross section is seen to be somewhat in error. These factors are calculated using the erroneous assumption that the differential cross sections remain fixed as temperature changes and that the temperature dependence is carried only in the change of the neutron spectrum with energy. Including the effects of temperature change on the differential cross section will change the g factors somewhat.

Calculations also were carried out for the values of $\epsilon = 11$, 14, and 17 meV for a number of temperatures. The influence of different values of ϵ was in the 0.1% or smaller range for temperatures from 20–600 K. Therefore the cross section displays only a very small dependence on chemical form. However, before attempting to interpret the low energy cross section with R matrix[13] or Adler-Adler analyses,[14] the measured data should be corrected for the phonon-transfer effect indicated on Figs. 2 and 3.

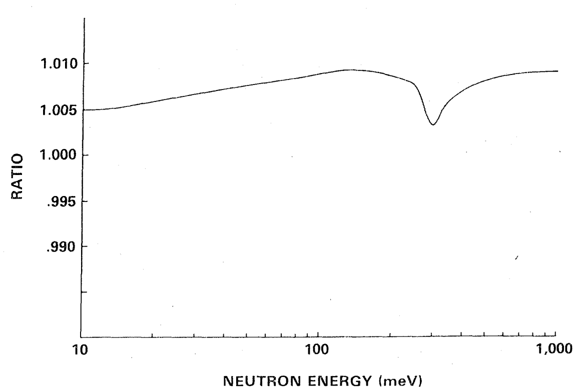

FIG. 5. The fractional phonon exchange effect for the fission cross section of ^{235}U is shown as calculated for Fig. 4.

Γ_n DEPENDENCE

The transmission of the s-wave potential barrier is often expressed in terms of the neutron wave number using the expression $T \alpha kK/(K^2+k^2) \simeq k/K$ where k and K are the wave numbers of the neutrons outside and inside of the potential barrier, respectively. Since a phonon carried a momentum of ω/v_a where ω is the circular frequency of the lattice oscillator and v_a is the speed of sound within the solid, the effective wave number k' operating in the barrier penetrability depends on the phonon transfer. Therefore the expression for conservation of momentum can be expressed in terms of the wave number as

$$\vec{k}' = \vec{k} - \sum_s (n_s - \alpha_s)\vec{\omega}_s/v_a - \vec{\Omega}\,, \tag{24}$$

where we assign a vector direction to each of the phonons created or absorbed from the lattice. The quantity $\vec{\Omega}$ is the momentum transferred to the crystal as a whole which, of course, carries no energy if the crystal is large. The vector sum $\sum_s \vec{\omega}_s/v_a$ is usually not colinear with k since the momentum balance is provided by $\vec{\Omega}$ although the vector sum and $\vec{\Omega}$ simultaneously may be colinear with $\vec{k}$.

The detailed question of the distribution over solid angle of the $\vec{\omega}_s$ and their sum is beyond the scope of this paper. However, an estimate of the influence of phonon transfer on neutron width can be obtained by assuming that both Ω and all $\vec{\omega}_s$ are always colinear with k. This assumption gives the maximum momentum transfer involving phonons for a given amount of energy transfer. In this case the vectors disappear from Eq. (24) and we have

$$k' = k - \sum_s (n_s - \alpha_s)\omega_s / v_a \,. \tag{25}$$

The wave number Ω is negligibly small compared to k since the crystal mass is so large compared to that of the neutron.

The dependence of neutron width on penetrability is generally written in terms of the reduced neutron width Γ_n^0 defined by the expression $\Gamma_n = \Gamma_n^0 (E \text{ eV}/1 \text{ eV})^{1/2}$ since $k = (2mE/\hbar^2)^{1/2}$. Therefore Γ_n can be written in terms of k using the expression $\Gamma_n = \Gamma_n^0 \hbar k/(2m)^{1/2}$, and using Eq. (25), Γ_n may be written including the influence of phonon transfer as

$$\Gamma_n = \Gamma_n^0 \hbar k'/(2m)^{1/2} = \Gamma_n^0 \sqrt{E}\left(1 - \sum_s (n_s - \alpha_s)\hbar\omega_s/(4E_a E)^{1/2}\right), \tag{26}$$

where E_a is the energy a neutron would have moving with the speed of sound within the material.

From Lamb we can then write that the expression for $W(E)$ including, in an approximate way, the effect of phonon exchange on the neutron width as

$$W(E) = \Gamma_\gamma \Gamma_n^0 E \times \sum_{\alpha_s} g_s \sum_{n_s} \frac{[1 - \rho_s/(4E_a E)^{1/2}] \,|(n_s|\exp(i\vec{k}\cdot\vec{\chi}_L)|\alpha_s)|^2}{(E - E_0 - \rho_s)^2 + \frac{1}{4}\Gamma^2]} , \tag{27}$$

where $\rho_s = \sum_s (n_s - \alpha_s)\hbar\omega_s$. Following Lamb's approach involving a δ function to perform the sums and appropriate averaging we obtain the expression

$$W(E) = \frac{\Gamma_\gamma \Gamma_n^0}{2\pi}\sqrt{E}\int_{-\infty}^{\infty} d\mu\, e^{g(\mu)} \int_{-\infty}^{\infty} \frac{e^{i\mu\rho}[1 - \rho/(eE_a E)^{1/2}]}{(E - E_0 - \rho)^2 + \frac{1}{4}\Gamma^2} . \tag{28}$$

The integral can be broken into two parts. The first integral yields the value for $W(E)$ of Eq. (23). The inner part of the second integral may be written

$$\int_{-\infty}^{\infty} \frac{e^{i\mu\rho}\rho\, d\rho}{(E - E_0 - \rho)^2 + \frac{1}{4}\Gamma^2} = \frac{2\pi}{\Gamma}(E - E_0 \mp \tfrac{1}{2} i\Gamma) \times \exp[i\mu(E - E_0 \mp \tfrac{1}{2} i\Gamma)] \tag{29}$$

with the minus sign for $\mu < 0$ and the plus sign for $\mu > 0$. Using the Einstein (δ function) approximation for the phonon frequency spectrum, the integral over μ can be carried out and when added

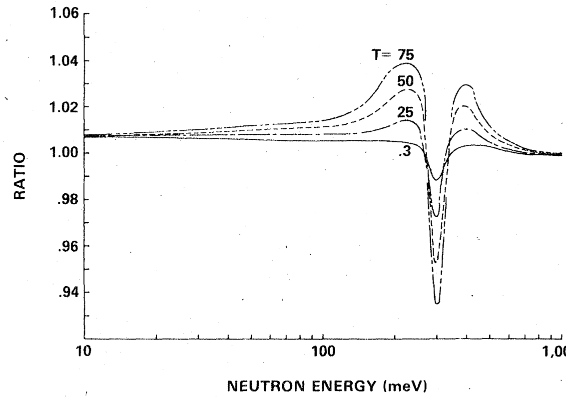

FIG. 6. Same as Fig. 2 with phonon dependence of Γ_n included.

to the first integral of Eq. (28), we find for $W(E)$

$$W(E) = \Gamma_\gamma \Gamma_n^0 \sqrt{E}\, e^{-b(2\alpha+1)} + \sum_{n=-\infty}^{\infty} \frac{(\alpha/\alpha+1)^{n/2} I(n)\{2b[\alpha(\alpha+1)]^{1/2}\}}{(E - E_0 + n\epsilon)^2 + \frac{1}{4}\Gamma^2} \times [1 + n\epsilon/(4EE_a)^{1/2}] , \tag{30}$$

where E_a is the energy of a neutron moving with the speed of sound in the sample material.

Calculations of Eq. (30) divided by the resonance denominator without Doppler broadening for ^{239}Pu and ^{235}U are shown in Figs. 6 and 7 using a speed of sound of 2900 m/sec. As with Eq. (23) the calculations appear to be essentially independent of modest changes in the value of ϵ. The temperature dependence is still present and effects at thermal are more noticeable than in Figs. 2 and 3.

Figures 8 and 9 show calculations of Eq. (30) divided by the Doppler broadened Breit-Wigner denominator for ^{239}Pu and ^{235}U, respectively. The effects at thermal are now amplified somewhat and the temperature dependence appears to be present throughout the energy region. Note that

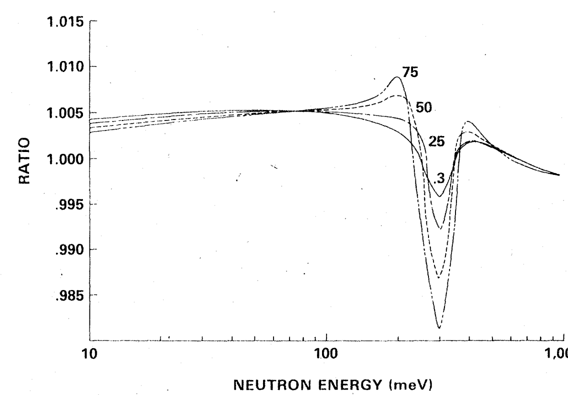

FIG. 7. Same as Fig. 3 with phonon dependence of Γ_n included.

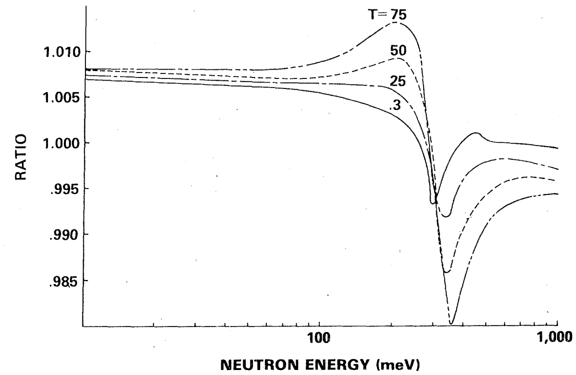

FIG. 8. Same as Fig. 4 with phonon dependence of Γ_n included.

the direction of the temperature dependence is opposite at low energies for ^{239}Pu and ^{235}U. This is a consequence of the resonance structure. For ^{235}U capture, one could expect a curve significantly different in shape.

The results of the calculation are summarized below:

(1) The magnitude of the phonon exchange effect without Γ_n phonon dependence is temperature dependent ranging in size from a few tenths of 1% at thermal energy for ^{235}U to ±5% for ^{239}Pu at 0.3 eV.

(2) The phonon exchange effect without Γ_n phonon dependence probably can be satisfactorily approximated for most purposes by applying the usual gas model.

(3) The inclusion of the phonon dependence on both Γ_n and the Breit-Wigner denominator results in temperature dependence for both ^{235}U and ^{239}Pu which follow the gas model approximation fairly well above 0.1 eV but much less well at thermal.

(4) Comparing Fig. 1 with Figs. 3, 5, 7, and 9, we see significant differences at thermal and subthermal energies indicating the inadequacy of the Einstein approximation at these very low energies.

These calculations therefore indicate that phonon transfer has effects at low energies which are small but probably not negligible. They serve to estimate the size of the effect, but do not take into account the details of the phonon spectrum. This limitation appears to be particularly important in the thermal and subthermal energy region. Since for capture and fission cross sections the effects in general will be different and perhaps of opposite sign, the effects in the capture to fission ratio, α, might be greater than in either fission or capture together.

These effects are small enough to have been overlooked in the past, but not too small to measure. It appears that they might be most easily detected by temperature dependent total cross section measurements. If measurable, they almost certainly will be of significance for reactor design. This work also suggests that theoretical and experimental studies on the liquid and gaseous phases would be interesting from a basic physics viewpoint and might have significant technological importance as well.

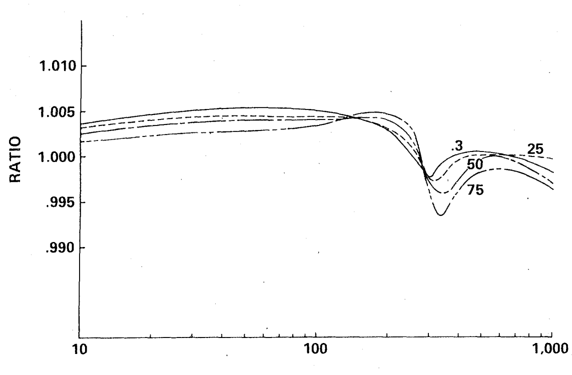

FIG. 9. Same as Fig. 5 with phonon dependence of Γ_n included.

CONCLUSIONS

The results show that reaction cross sections in general should depend on the chemical form of the sample, even though the effect is quite small, and on the sample temperature T. To the knowledge of the authors no such effect has been predicted or searched for before near thermal energies. This is somewhat surprising since the shape of cross sections at higher energy resonances has long been known to undergo changes with chemical composition and temperature and such effects have been carefully studied experimentally.

The size of the effects is not large but appear to be definitely measurable. Lemmel[15] points out some inconsistencies in existing measurements which perhaps hint at unforeseen factors influencing existing experiments. Very few measurements have been made with liquid or gaseous samples or on a sample as a function of temperature. An exception to the latter type of measurement is the integral reactor experiment in which, however, both the temperature of the sample and the neutron spectrum is changed. As Lemmel[15] points out, good agreement with predictions from differential measurements is often not achieved. With the existence of the proposed effect omitted from reactor calculations, one also would not expect thermal reactors or critical assemblies to behave in the way predicted. Effects of the same magnitude predicted for fission should also be expected in general for capture reactions on fissile or nonfissile isotopes. Therefore the capture-to-fission ratio α is also expected to exhibit some previously unexpected dependence on temperature. Although in this derivation a sum has been taken over all fission pairs, such a sum is not essential if one wishes to compute more details of the fission process. Since the mass distribution is known to change from resonance to resonance[16] and since phonon exchange can change the relative contributions of different resonances, one also expects measurable influences on the mass distribution. Likewise, since delayed neutrons are emitted from particular fission products, a change in the mass distribution might also change the delayed neutron yield per mg of fissile material. Such an effect has been reported by Lukens and Bramblett.[17]

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