it possible only to observe these deflections with an uncertainty of about 30%, the deflections were of the correct direction and appeared to be of the correct magnitude expected from the magnitude of the persistent angular momentum present and the rate of warming or cooling.

It is worthwhile considering the reversible temperature dependence of the persistent angular momentum from the viewpoint of the elementary-excitation model of Landau.[11] Consider a persistent current in a sphere at rest at absolute zero where the entire fluid is superfluid and has angular momentum **L**. Following an argument presented by Feynman,[14] as the temperature is increased from absolute zero thermal excitations appear which, in order to have zero average group velocity and remain in equilibrium with the stationary walls, have an average angular momentum which opposes and cancels part of the **L** possessed by the fluid background. Thus, a net torque must be exerted on the liquid to create these excitations which lowers the total angular momentum of the liquid. When the temperature is lowered again, these excitations disappear, giving up their angular momentum to the walls and thus removing the angular momentum which tended to cancel **L**. Hence, as the temperature returns to absolute zero, the total fluid angular momentum returns to the original **L**. The temperature variation of the persistent angular momentum can thus be interpreted as being due to the creation and destruction of excitations superimposed on a fluid background which remains in motion.

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Brownian Motion of a Heavy Impurity in Harmonic Lattices

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Starting from the well-known expression for the frequency spectrum of a mass impurity in a harmonic lattice, the velocity autocorrelation function for a heavy impurity has been calculated without referring to any particular model. Using then general arguments, the postulates of Langevin's equation have been briefly discussed.

1. INTRODUCTION

The motion of a heavy impurity in harmonic lattices has been studied by Rubin[1,2] and others[3] with a view to justifying the postulation of a phenomenological equation of the Langevin type. In particular, Mazur[3] has discussed general conditions under which a heavy mass in a harmonic lattice will perform Brownian motion. The purpose of this paper is to show how, starting from the well-known result for the spectral function of an impurity in a harmonic lattice, one can derive in a simple and straightforward manner the velocity autocorrelation function for the impurity. We have derived exact expressions for that function in the heavy-mass limit for one-, two-, and three-dimensional harmonic lattices. In the one-dimensional case, using the fluctuation-dissipation theorem of statistical physics, the postulates of the Langevin equation have been rigorously justified.

2. MATHEMATICAL FORMULATION

The real part of the velocity autocorrelation is expressed in terms of the frequency distribution function $f(\omega)$ as follows[4]:

$$\mathrm{Re}\langle \mathbf{v}(t)\cdot\mathbf{v}(0)\rangle = \frac{\nu h}{2M'}\int_0^\infty \omega \coth(h\omega/2k_BT) f(\omega)\cos(\omega t)d\omega\,, \quad (1)$$

where ν stands for the dimensionality of the lattice, and the other symbols have their usual meaning.

For a harmonic solid with cubic symmetry, $f(\omega)$ for an isolated localized impurity has been given by Waller[5]

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[1] R. J. Rubin, J. Math. Phys. **1**, 309 (1960).

[2] R. J. Rubin, J. Math. Phys. **2**, 373 (1961).

[3] P. Mazur, in *Statistical Mechanics of Equilibrium and Non-Equilibrium*, edited by J. Meixner (North-Holland Publishing Co., Amsterdam, 1956), p. 63; P. Mazur and E. Braun, Physica **2**, 1973 (1964); H. Nakazawa, Progr. Theoret. Phys. (Kyoto) Suppl. **36**, 176 (1966).

[4] See, for instance, A. Rahman, K. S. Singwi, and A. Sjölander, Phys. Rev. **126**, 986 (1962).

[5] I. Waller, Arkiv Fysik **24**, 495 (1963).

and others[6] using a Green's-function technique under the assumption that the force constants remain unchanged. The expression is

$$f(\omega)=\frac{2(1+\eta)\omega g(\omega^2)}{[1+\eta\omega^2 F(\omega^2)]^2+[\pi\eta\omega^2 g(\omega^2)]^2}, \tag{2a}$$

where $\eta=(M'-M)/M$ and

$$F(\omega^2)=P\int\frac{g(\omega'^2)d\omega'^2}{\omega^2-\omega'^2}. \tag{2b}$$

Here P stands for the principal value, $g(\omega^2)$ gives the density of phonon states between ω^2 and $\omega^2+d\omega^2$, and M' and M denote the masses of an impurity and a host atom, respectively.

Substituting Eq. (2a) in Eq. (1), one gets the following expression for the velocity autocorrelation function for the impurity atom:

$$\Phi(t)\equiv\mathrm{Re}\langle\mathbf{v}(t)\cdot\mathbf{v}(0)\rangle$$
$$=\frac{\nu h}{M}\int_0^\infty\frac{\omega^2 g(\omega^2)\coth(\hbar\omega/2k_BT)\cos(\omega t)d\omega}{[1+\eta\omega^2F(\omega^2)]^2+[\pi\eta\omega^2 g(\omega^2)]^2}. \tag{3}$$

Three cases of a one-, two-, and three-demensional (1D, 2D, and 3D) host lattice will be considered separately, and we shall let $M'\to\infty$. We first notice that the integration above extends only up to a certain maximum frequency ω_L. Of particular importance is the behavior of $g(\omega^2)$ for $\omega\to 0$ and, therefore, we state below its general form for small ω in the three cases[7]:

$$\begin{aligned} g(\omega^2) &\to a/\omega \quad &&\text{(1D lattice)}\\ &\to b &&\text{(2D lattice)}\\ &\to c\omega &&\text{(3D lattice)}, \end{aligned} \tag{4a}$$

where a, b, and c are constants which are determined by interatomic force constants. For small ω one can show that

$$\begin{aligned} F(\omega^2) &\to a' &&\text{(1D lattice)}\\ &\to -b\ln(b'/\omega^2) &&\text{(2D lattice)}\\ &\to -c' &&\text{(3D lattice)}, \end{aligned} \tag{4b}$$

a', b', and c' being constants.

The behavior of $g(\omega^2)$ near $\omega=\omega_L$ is also known in general and is stated below[8]:

$$\begin{aligned} g(\omega^2) &\to \mathrm{const}(\omega_L^2-\omega^2)^{-1/2} &&\text{(1D lattice)}\\ &\to \mathrm{const}\neq 0 &&\text{(2D lattice)}\\ &\to \mathrm{const}(\omega_L^2-\omega^2)^{1/2} &&\text{(3D lattice)}. \end{aligned} \tag{4c}$$

3. ONE-DIMENSIONAL LATTICE

Putting $x=\omega\eta/\omega_L$ in Eq. (3) and splitting the range of integration into two parts, we have

$$\Phi(t)=\frac{\hbar\omega_L}{M\eta}\left\{\int_0^{x_0}+\int_{x_0}^{\eta}\frac{(\omega_Lx/\eta)^2 g(\omega_L^2x^2/\eta^2)\coth(\hbar\omega_Lx/2k_BT\eta)\cos(\omega_Lxt/\eta)dx}{[1+(\omega_L^2x^2/\eta)F(\omega_L^2x^2/\eta^2)]^2+[(\pi\omega_L^2x^2/\eta)g(\omega_L^2x^2/\eta^2)]^2}\right\}, \tag{5}$$

where we shall let $x_0\to\infty$, when $\eta\to\infty$, such that $x_0/\eta\to 0$. We can write $x_0=\eta^{1-\alpha}$, where α is any arbitrary positive number between zero and unity.

After making use of the stated properties of $g(\omega^2)$ and $F(\omega^2)$ in evaluating the first integral in Eq. (5), we obtain

$$\frac{2k_BTa\omega_L}{M\eta}\int_0^{x_0}\frac{\cos(\omega_Lxt/\eta)dx}{1+(\pi\omega_Lax)^2}$$
$$=\frac{k_BT}{M'}[\exp(-t/\pi\eta a)+O(\eta^{-1+\alpha})]. \tag{6}$$

The second integral in Eq. (5) is found to be smaller than

$$\frac{\hbar\omega_L}{M\eta}\int_{x_0}^{\eta}\frac{x\coth(\hbar\omega_Lx/2k_BT\eta)dx}{\pi^2\omega_L^2x^3g(x^2\omega_L^2/\eta^2)}=\frac{1}{M'}O(\eta^{-1+\alpha}), \tag{7}$$

where we have used the fact that in the range of integration the maximum of $[x\coth(\hbar\omega_Lx/2k_BT\eta)]$ is proportional to η, and that $(\omega_Lx/\eta)g(\omega_L^2x^2/\eta^2)$ has a positive minimum which is independent of η.

Hence, the final result is

$$\Phi(t)=\frac{k_BT}{M'}[\exp(-t/\pi a\eta)+O(\eta^{-1+\alpha})] \tag{8}$$

for $\eta\to\infty$.

Now, if we let $t\to\infty$ in such a way that t/η is finite for $\eta\to\infty$, then

$$\Phi(t)\to\frac{k_BT}{M'}\exp(-t/\pi a\eta). \tag{9}$$

This result is the same as obtained by Mazur,[3] and is also the same as obtained by Rubin[2] in his nearest-neighbor-interaction model if we identify his $2\gamma^{1/2}$ with our $1/\pi a$, where γ is the force constant.

4. TWO-DIMENSIONAL LATTICE

In this case, $\Phi(t)$ is given by Eq. (5) except for a factor 2 in front of it. Proceeding in the same way as above, it is easy to show that the second integral in

[6] R. J. Elliott and D. W. Taylor, Proc. Phys. Soc. (London) **83**, 189 (1964); M. Yu. Kagan and A. P. Zernov, Zh. Eksperim. i Teor. Fiz. **50**, 1107 (1966) [English transl.: Soviet Phys.—JETP **23**, 737 (1966)].

[7] A. A. Maradudin, E. W. Montroll, and G. H. Weiss, *Theory of Lattice Dynamics in the Harmonic Approximation* (Academic Press Inc., New York, 1963), p. 56.

[8] L. van Hove, Phys. Rev. **89**, 1189 (1953).

Eq. (5) is again smaller than

$$(\text{const}/M')\eta^{-1+\alpha}, \quad (0<\alpha<1) \tag{10}$$

where we have used the fact that $g(\omega_L^2x^2/\eta^2)$ is different from zero throughout the whole integration region.

In the first integral of Eq. (5), $x/\eta \to 0$ as $\eta\to\infty$ and we can use the expressions given in Eqs. (4a) and (4b). The integral can be brought into the form

$$\frac{4k_BT}{\pi M\eta^2}\int_0^{\omega_L(\pi b)^{1/2}x_0}\frac{\rho\cos(\rho t/(\pi b)^{1/2}\eta)d\rho}{[1-(\rho^2/\pi\eta)\ln(\pi bb'\eta^2/\rho^2)]^2+[\rho^2/\eta]^2}. \tag{11}$$

In the integrand, the denominator vanishes for $\rho=\epsilon\eta e^{i\delta}$, with ϵ and δ determined by the equations,

$$\begin{aligned}\epsilon^2\eta&=\pi[\ln(\pi bb'/\epsilon^2)]^{-1},\\ \delta&=\tfrac{1}{2}\pi[\ln(\pi bb'/\epsilon^2)]^{-1},\end{aligned} \tag{12}$$

for $\eta\to\infty$. We notice that as $\eta\to\infty$, $\epsilon\to 0$ and $\delta\to 0$. Also

$$\epsilon\eta=\pi[\epsilon\ln(\pi bb'/\epsilon^2)]^{-1}\to\infty$$

and

$$\epsilon\eta\delta=\pi^2/2\epsilon[\ln(\pi bb'/\epsilon^2)]^2\to\infty\,.$$

Changing now the integration variable to

$$y=\rho-\epsilon\eta\,,$$

we get the expression

$$\frac{4k_BT}{\pi M\eta^2}\int_{y_1}^{y_2}\frac{(\epsilon\eta+y)\cos[(\epsilon\eta+y)\tau/\eta]dy}{\{1-[(\epsilon\eta+y)^2/\pi\eta]\ln[\pi bb'\eta^2/(\epsilon\eta+y)^2]\}^2+(\epsilon\eta+y)^4/\eta^2}, \tag{13}$$

where $\tau=t/(\pi b)^{1/2}$, $y_1=-\epsilon\eta$, and $y_2=\omega_L(\pi b)^{1/2}x_0-\epsilon\eta$.

We further split the integration range in three regions, $(-\epsilon\eta, -y_0)$, $(-y_0, y_0)$, and $[y_0, \omega_Lx_0(\pi b)^{1/2}-\epsilon\eta]$, such that $\epsilon\eta\gg|y_0|\gg\epsilon\eta\delta$. When integrating from $-y_0$ to y_0, we can expand in powers of y, and using Eq. (12) the integral can be written as

$$\frac{4k_BT}{\pi M\eta^2}\int_{-\infty}^{+\infty}\frac{(\epsilon\eta+y)\cos[(\epsilon\eta+y)\tau/\eta]dy}{(\epsilon y/\delta)^2+(\epsilon^2\eta)^2}$$

for $\eta\to\infty$. All the correction terms arising from other regions of integration are shown to be negligible and we arrive at the result

$$\Phi(t)=\frac{2k_BT}{M'}[\exp(-\epsilon\tau\delta)\cos(\epsilon\tau)+O\{\ln(\pi bb'/\epsilon^2)^{-1+\alpha}\}], \quad (0<\alpha<1). \tag{14}$$

For the nearest-neighbor-interaction model, $\tau=2\gamma^{1/2}t$, since $b=1/4\pi\gamma$ is the force constant. This agrees with the result of Rubin.[1]

5. THREE-DIMENSIONAL LATTICE

In this case $\Phi(t)$ is again given by Eq. (5) except for a factor 3 in front of it. Proceeding in the same way as before, it is straightforward to show that the first integral can be written as

$$\frac{6k_BT\omega_L^3c}{M\eta^3}\int_0^{x_0}\frac{x^2\cos(\omega_Lxt/\eta)dx}{[1-\omega_L^2x^2c'/\eta]^2+[\pi c\omega_L^3x^3/\eta^2]^2},$$

where c and c' are the constants appearing in Eqs. (4a) and (4b) and where we should let $x_0\to\infty$. The integral can be evaluated by the method of residue calculus and the result is

$$\frac{3k_BT}{M'}[\exp(-\tfrac{1}{2}\beta t)\{\cos(\omega_0t)-(\beta/2\omega_0)\sin(\omega_0t)\} +O(1/x_0)], \tag{15}$$

where $\beta=\pi c/c'^2\eta$ and $\omega_0=(c'\eta)^{-1/2}$. The second integral of Eq. (5) can be shown to be smaller than

$$\text{const}\frac{\eta}{M'}\int_{x_0}^{\eta}\frac{dx}{x^3g(\omega_L^2x^2/\eta^2)}=\frac{1}{M'}O(\eta^{-1+\alpha}), \tag{16}$$

with $0<\alpha<1$.

It should be noted that in the above integral, $g(\omega_L^2x^2/\eta^2)$, vanishes at the upper limit. Hence in its evaluation it has been split into two parts and the explicit dependence of $g(\omega^2)$ near the upper limit stated in Eq. (4c) has been used.

Combining Eqs. (15) and (16), we get finally

$$\Phi(t)=\frac{k_BT}{M'}[\exp(-\tfrac{1}{2}\beta t)\{\cos(\omega_0t)-(\beta/2\omega_0)\sin(\omega_0t)\} +O(\eta^{-1+\alpha})]. \tag{17}$$

In order to compare this result with that of Rubin,[2] one has to evaluate the constants c' and c in the nearest-neighbor-interaction model. In this case, c is given by

$$c=(4\pi^2\gamma^{3/2})^{-1},$$

γ being the force constant. It remains to be shown that c' is identical to [see Eq. (25) in Ref. 1]

$$\zeta(0,0)=\lim_{p\to 0}\int_0^\infty\exp(-p^2u)f(u)du \tag{18}$$

in Rubin's notation, and where

$$f(u)=\exp(-b\gamma u)[J_0(2i\gamma u)]^3,$$

J_0 being the Bessel function of zero order. Since [see Eq. (3.2.18a) in Ref. 7]

$$g(\omega^2)=\frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty}\exp(u\omega^2)f(u)du, \tag{19}$$

we get by inversion

$$f(u)=\int_0^\infty e^{-u\omega^2}g(\omega^2)d\omega^2\,, \tag{20}$$

which when substituted in Eq. (18) gives

$$\zeta(0,0)=\int_0^\infty \frac{g(\omega^2)d\omega^2}{\omega^2}. \tag{21}$$

This is obviously the same as c' defined in Eq. (4b) and in connection with Eq. (2).

According to Rubin,[2] one has

$$\zeta(0,0)=\zeta_0/4\gamma\,,$$

where $\zeta_0\approx 1.019$. β and ω_0 appearing in Eq. (17) are now given by

$$\beta=4\gamma^{1/2}/\pi\zeta_0{}^2\eta$$

and

$$\omega_0=2\gamma^{1/2}(\zeta_0\eta)^{-1/2}.$$

With these values of β and ω_0, Eq. (17) is the same as given by Rubin.[2]

6. REMARKS

In the foregoing sections we have seen that the velocity autocorrelation of an infinitely heavy impurity varies as an exponential only in the one-dimensional case. In two and three dimensions it varies like that of a damped harmonic oscillator, where the damping tends to zero with increasing mass.

In order to justify the postulates of the phenomenological equation of Langevin, we shall consider the one-dimensional case only. It is straightforward to show that the equation of motion for the impurity can be written in the following form[3]:

$$\dot{p}(t)+\gamma(t)p(t)=E(t)\,, \tag{22}$$

where $p(t)$ is the momentum of the heavy impurity, and $E(t)$ represents the force exerted on it by the surrounding atoms, which depends linearly on the initial positions and momenta of the atoms of the host lattice but is independent of the impurity itself. $E(t)$ is thus a sum of Gaussian stochastic variables, and is, hence, itself a Gaussian stochastic variable.[9] $\gamma(t)$ is a time-dependent function which does not contain any of the particle variables. From Eq. (22), one directly obtains by multiplying with $p(0)$ and taking a statistical average the following equation for the velocity autocorrelation function:

$$\dot{\Phi}(t)+[\gamma(t)/M']\Phi(t)=0. \tag{23}$$

However, we have shown that

$$\Phi(t)=(k_BT/M')\exp(-t/\pi a\eta)$$

for $\eta\rightarrow\infty$, and this implies that $\gamma(t)$ is constant in time. If we now use this result in conjunction with the fluctuation-dissipation theorem,[10] we conclude that the correlation function $\langle E(t)E(0)\rangle$ is proportional to a δ function, i.e., $E(t)$ has a white spectrum. Therefore, for the one-dimensional case all the postulates of Langevin's equation are rigorously justified.

It may be noted that in the case of two and three dimensions, $\gamma(t)$ is not a constant and, therefore, $E(t)$ does not have a white spectrum. $E(t)$ still depends linearly on the initial positions and momenta of the host lattice atoms, and hence it has a Gaussian distribution at any time.

Lastly, we might remark that Eq. (2) may be used as a reasonable approximation to find the velocity autocorrelation function of an impurity in a liquid, provided a suitable $f(\omega)$ characteristic of a liquid is used.

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[9] We have here followed the argument given by Mazur in Ref. 3.

[10] R. Kubo, in *Many-Body Theory*, edited by R. Kubo (W. A. Benjamin and Company, Inc., New York, 1966), Part I, p. 1.