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Spinodal temperatures for macroscopic density fluctuations in Pd–H: I. Eigenvalues of hydrogen density modes for a thin-disc geometry

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Abstract. The elastic eigenvalues associated with hydrogen density fluctuations near the critical point of a metal–hydrogen system have been evaluated approximately for the case of a thin-disc geometry, with a small but finite thickness.

1. Introduction

In metal–hydrogen systems which display an analogue of a liquid–gas transition (e.g. Pd–H and Nb–H), the hydrogen interstitials can be thought of as lattice gas particles [1–4] with a very long-range interaction. There is a coherency regime in the phase diagram where the correlation length is of the order of the sample size. The hydrogen density in this region tends not to form abrupt gradients because of the high stress energy involved, but varies in long-range fluctuations whose form and energy is determined by the geometry and the conditions at the surface [5–7]. These fluctuations are called macroscopic density modes, and are the fundamental thermodynamic fluctuations of the system. To determine the free energy, and hence the thermodynamics, of the metal–hydrogen system at the critical point it is necessary to specify the characteristic energies of the critical fluctuations.

Experiment has determined the presence of some of these modes in various sample geometries of Nb–H; by neutron radiography [8] in short cylinders, the Gorsky effect in foils [9], and the broadening of x-ray peaks [10] in thin discs. It is difficult to determine the exact excitation energies and spinodal temperatures characteristic of each fluctuation. Exact calculations for a spherical system and for the general ellipsoidal system [5, 11] have been done, as well as for an exactly two-dimensional system [12, 13]. It is worth commenting on the comparison with the (exact) analytical calculation for the general ellipsoid done by Maier-Bötzel and Wagner, since a sufficiently oblate ellipsoid can be thought of as a disc for many purposes. The differences between the boundary conditions of a disc system, with two flat planar surfaces forming part of the surface, and those of an ellipsoid, with a smooth differentiable surface everywhere, are manifested in the form of the eigenfunctions and also in the values for the eigenvalues [11] and in equations (4.1) and (4.2) of this work.

Some experiments on thin samples of Pd–H [14, 15] are being done on samples with a small but finite thickness, so the possibility has to be considered that the characteristic

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modes might be changed from the exactly two-dimensional case considered in [12, 13]. To interpret the results from such an experiment one requires accurate estimates of the spinodal temperatures for the modes in a sample of small but finite thickness, hence the motivation for the calculation presented here. The results of this calculation should, of course, be compatible with the details for the exactly two-dimensional system in the limit of infinitesimal thickness.

This paper details the calculation of the characteristic energies of deformation of a particular geometry, the thin disc, assuming the elastic constants of the underlying matrix are known. Although the essentials of this calculation hold for coherent metal–hydrogen systems generally, specific parameters and conditions pertinent to the palladium–hydrogen system are used in this development.

An exact solution for the three-dimensional elastic problem in cylindrical coordinates involves two coupled, non-orthogonal, Fourier–Bessel series. The elastic modes have therefore been approximated by a sum of spherical harmonics and the largest corrections due to edge effects modelled by a Bessel series.

The form most often used for the elastic energy in metal–hydrogen systems is [5]

$$E_{el} = \int \{\tfrac{1}{2}\varepsilon_{\alpha\beta}(r)C_{\alpha\beta\mu\nu}(r) - \rho(r)P_{\alpha\beta}\varepsilon_{\alpha\beta}(r)\}\,dr \tag{1.1}$$

where the strain $\varepsilon_{\alpha\beta}$ and elastic constants $C_{\alpha\beta\mu\nu}$ are defined in the host lattice, ρ is the density of hydrogen atoms, and $P_{\alpha\beta}$ is the force dipole tensor, which is a measure of the interaction of the lattice with a hydrogen defect. The stress tensor is given by

$$\sigma_{\alpha\beta} = C_{\alpha\beta\mu\nu}\varepsilon_{\mu\nu}(r) - P_{\alpha\beta}\rho(r) \tag{1.2}$$

and must satisfy the equations of equilibrium in a stress-free system

$$\partial_\alpha \sigma_{\alpha\beta} = 0 \tag{1.3}$$

$$n_\alpha \sigma_{\alpha\beta} = 0 \tag{1.4}$$

at the boundary, with n_α the normal to the surface. The energy may be rewritten

$$E_{el} = \tfrac{1}{2}\int \varepsilon_{\alpha\beta}P_{\alpha\beta}\rho(r)\,dr. \tag{1.5}$$

Since the Hamiltonian is linear in the density of hydrogen, the associated strain is some linear functional of the density distribution [5]

$$P_{\alpha\beta}\varepsilon_{\alpha\beta} = \int W(r, r')\rho(r')\,dr'. \tag{1.6}$$

It is the eigenfunctions $\varphi_l(r)$ of the kernel W that are the normal elastic modes of the system. The energy eigenvalues, defined by

$$\int W(r, r')\varphi_l(r')\,dr' = \omega_l\varphi_l(r) \tag{1.7}$$

can be used to express the elastic energy of the system simply as

$$E_{el} = -\tfrac{1}{2}\sum_l \omega_l\rho_l^2 \tag{1.8}$$

where ρ_l is the amplitude of the density mode

$$\rho_l = \frac{1}{\sqrt{V}}\int \rho(r)\varphi_l(r)\,dr. \tag{1.9}$$

Comparison of equation (1.5) and (1.8) shows that, when only one mode is excited,

$$P_{\alpha\beta}\varepsilon_{\alpha\beta} = \omega_l \varphi_l(r). \tag{1.10}$$

The cubic symmetry of the palladium lattice forces the force dipole tensor to be diagonal, with all components equal. In addition, it is convenient to make the assumption of isotropy in order to obtain a solution. Anisotropy in the lattice has the effect of distorting the modes obtained from the isotropic equations [16]. With the usual Lamé constants the total stress for one mode may be written

$$\sigma_{\alpha\beta} = \lambda\delta_{\alpha\beta}\nabla \cdot \boldsymbol{u} + \mu(\partial_\alpha u_\beta + \partial_\beta u_\alpha) + \mathbf{P}\delta_{\alpha\beta}\rho(r). \tag{1.11}$$

This may be thought of as a stress-free system with a modified Lamé constant [11]

$$\lambda' = \lambda - \mathbf{P}^2/\omega. \tag{1.12}$$

One looks for sustainable deformations in the absence of external stresses. The corresponding value for λ' can then be used to work out the eigenvalue for the mode via equation (1.12). The characteristic energy of the fluctuation is related to the spinodal temperature as shown in section 5.

2. Eigenvalues of a thin isotropic disc

The Papkovich–Neuber formalism [5] for the displacement is used [17]:

$$\boldsymbol{u} = \nabla(\Phi + \boldsymbol{\rho} \cdot \boldsymbol{\Psi}) - 2(2 - \gamma)\boldsymbol{\Psi} \tag{2.1}$$

where $\gamma = \lambda/(\lambda + \mu)$, Φ and $\boldsymbol{\Psi}$ are harmonic functions, and $\boldsymbol{\rho} = \boldsymbol{r} + \boldsymbol{z}$ is the position vector in spherical polars, $\boldsymbol{r}$ is the radial vector in cylindrical coordinates. The spherical polars are used only for convenience when writing down the potential; the system to be studied is a cylindrical one.

It is convenient to expand Φ and $\boldsymbol{\Psi}$ in terms of spherical harmonics, namely

$$\Phi = \sum_{n=0}^{s+1} A_n \rho^{l+1+2n} P^l_{l+1+2n}(\cos\theta)\cos l\varphi \tag{2.2}$$

$$\begin{aligned}\boldsymbol{\Psi} = \sum_{n=0}^{s+1} [&C_n P^{l+1}_{l+2n}(\cos\theta)(\hat{\boldsymbol{r}}\cos l\varphi + \hat{\boldsymbol{\varphi}}\sin l\varphi) \\ &+ D_n P^{l-1}_{l+2n}(\cos\theta)(\hat{\boldsymbol{r}}\cos l\varphi - \hat{\boldsymbol{\varphi}}\sin l\varphi)]\rho^{l+2n} \\ &+ E_n \rho^{l+2n} P^l_{l+2n}(\cos\theta)\cos l\varphi\hat{\boldsymbol{z}}\end{aligned} \tag{2.3}$$

where (ρ, θ, φ) are spherical coordinates, and the highest power of r, the cylindrical radius vector, in the dilation is r^{l+2s}.

These potentials describe displacements of odd parity along the z-axis; similar expressions can be obtained for variations of even partity:

$$\Phi = \sum_{n=0}^{s+1} A_n \rho^{l+2n} P^l_{l+2n}(\cos\theta)\cos l\varphi \tag{2.4}$$

$$\begin{aligned}\boldsymbol{\Psi} = \sum_{n=0}^{s+1} &(C_n P^{l+1}_{l+2n-1}(\hat{\boldsymbol{r}}\cos l\varphi + \hat{\boldsymbol{\varphi}}\sin l\varphi) + D_n P^{l-1}_{l+2n-1}(\cos\theta)(\hat{\boldsymbol{r}}\cos l\varphi \\ &- \hat{\boldsymbol{\varphi}}\sin l\varphi))\rho^{l+2n-1} + E_n \rho^{l+2n-1} P^l_{l+2n-1}(\cos\theta)\cos l\varphi\hat{\boldsymbol{z}}.\end{aligned} \tag{2.5}$$

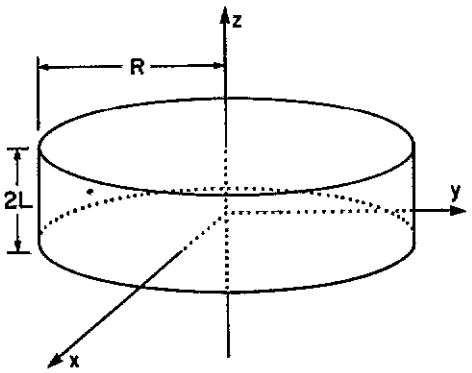

Figure 1. Geometry and axes for the system considered. The ratio L/R is small relative to unity.

Note first, that in either case the dilation Δ is given by

$$\Delta = 2(\gamma - 1)\nabla \cdot \boldsymbol{\Psi}. \tag{2.6}$$

The equations of equilibrium are automatically satisfied for zero-body forces in the interior of the system. The boundary conditions have two forms: at the flat surface of the disc, $z = \pm L$ (see figure 1),

$$\lambda\Delta = 2\mu\partial_z u_z = 0 \tag{2.7}$$

$$\partial_\varphi u_z + \partial_z u_\varphi = 0. \tag{2.8}$$

At the circular peripheral boundary, $r = R$, the conditions are

$$\lambda\Delta + 2\mu\partial_r u_r = 0 \tag{2.9}$$

$$\partial_r u_\varphi + (\partial_\varphi / r)u_r - (u_\varphi / r) = 0. \tag{2.10}$$

The shear stress σ_{rz} will be neglected on both surfaces. Using equation (1.5),

$$\Delta E_{el} = -\tfrac{1}{2}\int_V \mathbf{P}\rho(r)\varepsilon'_{\alpha\alpha}\,\mathrm{d}r \tag{2.11}$$

where $\varepsilon'_{\alpha\alpha}$ is the change in strain in this approximation. By St Venant's principle this is localized near the surface in question; but then one may expect the strain caused by a shear force to have zero dilation. Further discussion of some of the approximations necessary for solutions in three dimensions can be found in Sokolnikov [18].

3. Details of the calculation

The case of modes of odd parity in z will be treated first. In terms of the expression for $\mathbf{u}$ the boundary conditions at the flat surfaces become

$$0 = -\gamma\nabla \cdot \boldsymbol{\Psi} + \partial_z^2\Phi + \partial_z^2(r\Psi_r + z\Psi_z) - 2(2 - \gamma)\,\partial_z\Psi_z \tag{3.1}$$

$$0 = \partial_\varphi\partial_z\Phi + \partial_\varphi\partial_z(r\Psi_r + z\Psi_z) - (2 - \gamma)(\partial_\varphi\Psi_z + \partial_z\Psi_\varphi). \tag{3.2}$$

There are $2s + 2$ linear equations in the $4s + 4$ coefficients of the various powers of r, which now separate out. Successive terms couple Legendre polynomials, the power of r decreasing in each case, the last term involving all the terms in the potentials (2.2) and (2.3).

The $2s + 2$ boundary conditions at $r = R$ can be satisfied exactly for all but one power of z, there being remnant terms in the powers of $r^{l-2}z^{2s+3}$, so that a Fourier–Bessel series needs to be added to complete the solution.

Without considering the corrections needed to balance the terms of highest order in z

$$0 \approx (\gamma + 2s + 2)\Delta(r). \tag{3.3}$$

Choosing the right-hand bracket to be zero causes all boundary conditions to be satisfied without coupling between different terms in the series. The approximate solution for the eigenvalue (neglecting the remnant terms at the peripheral boundary) is then

$$\gamma \approx -2(s + 1). \tag{3.4}$$

The effect of the remnant forces is to induce additional strains decreasing away from the edge. It is appropriate to expand the correction to the potentials in terms of the modified Bessel functions

$$\Phi' = \sum_k F_k I_l(kr) \sin kz \cos l\varphi \tag{3.5}$$

with wave vectors k defined by

$$k = (n + \tfrac{1}{2})\pi/L \tag{3.6}$$

where n is an integer. We then have

$$\Psi' = \sum_k [G_k(\hat{r}\cos l\varphi - \hat{\varphi}\sin l\varphi)I_{l-1}(kr) + H_k(\hat{r}\cos l\varphi + \hat{\varphi}\sin l\varphi)I_{l+1}(kr)]\sin kz. \tag{3.7}$$

With the values of k as chosen above, there is only one equation to be satisfied at the boundary $z = \pm L$;

$$0 = \sum_k \sin kL[k^2 I_l(kR)F_k + G_k(\gamma k I_l(kR) + k^2 R I_{l-1}(kR)) + H_k(\gamma k I_l(kR) + k^2 R I_{l+1}(kR))]. \tag{3.8}$$

Note that as the stress decreases exponentially away from the circular boundary, it is sufficient to satisfy the equations at $r = R$.

The effect of the additional stresses will be felt in the elastic eigenvalue, so that the approximate form of γ given above will have a correction term. We make the *ansatz* that the next lowest power of z is also obtained from the series. Then

$$\gamma \approx -2\left(s + 1 + \frac{l(l-1)}{2(s+1)}\frac{L^2}{R^2} + \ldots\right) \tag{3.9}$$

The values for the elastic energy eigenvalue are obtainable from this once the elastic constants for the (isotropic) matrix are known.

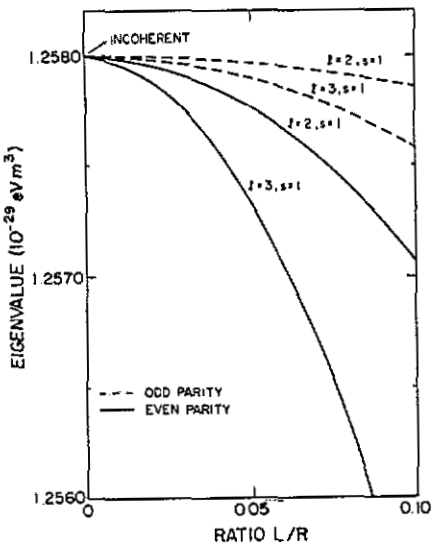

Figure 2. Variation of the eigenvalue for the first four modes with non-trivial variation.

The even parity choice of potentials yields another set of linear equations in descending powers of r and z. Another Fourier–Bessel series is needed to deal with boundary effects at $r = R$. The lowest order value for the constant γ is

$$\gamma \approx -2(s+1). \tag{3.10}$$

A Fourier–Bessel series must be used to correct for the edge effects; the next correction to the eigenvalue then becomes

$$\gamma \approx -2\left[s+1+\frac{l(l-1)}{(s+1)(2s+1)}\left(2s+3-\frac{2l}{l+2s}\right)\frac{L^2}{R^2}+\ldots\right]. \tag{3.11}$$

4. Summary

The geometric constant γ can be estimated for a thin disc to have the form (3.9) or (3.11) depending on the parity of the deformation in the axial direction. This gives for the elastic eigenvalue

$$\omega_{ls} = \mathbf{P}^2 \Big/ \left(\lambda + \frac{\gamma}{\gamma - 1}\mu\right) \approx \mathbf{P}^2\left[\lambda + \mu\left(\frac{2s+2}{2s+3} + \frac{l(l-1)}{(s+1)(2s+3)^2}\frac{L^2}{R^2}\right)\right]^{-1} \tag{4.1}$$

for the odd parity modes, and

$$\omega_{ls} \approx \mathbf{P}^2\left\{\lambda + \mu\left[\frac{2s+2}{2s+3} + \frac{l(l-1)}{(s+1)(2s+1)(2s+3)}\left(2s+3-\frac{2l}{l+2s}\right)\frac{L^2}{R^2}\right]\right\}^{-1} \tag{4.2}$$

for the even parity modes. These values are shown in figures 2 and 3 as a function of L/R

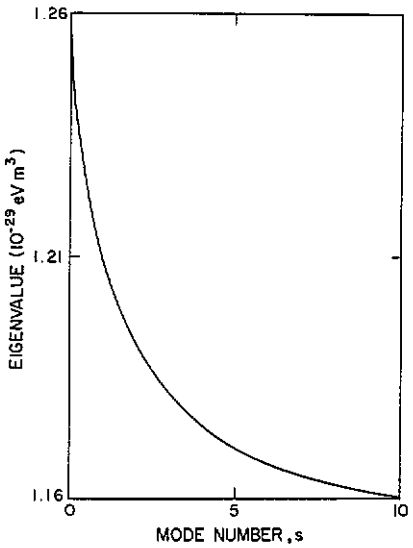

Figure 3. Eigenvalue versus s for $l = 0$, both even and odd parity modes.

and s respectively and in figure 4 the eigenvalues are given as a function of l for one particular (low value) choice of s.

With each macroscopic mode may be associated a spinodal curve, at which the energy required to excite the mode goes to zero. With the non-elastic interaction terms represented by a Landau local free energy,

$$F_0/V = kT(E_0 + \tfrac{1}{2}\alpha\rho^2 + \tfrac{1}{4}\beta\rho^4 + \ldots). \tag{4.3}$$

The coefficients in this expansion are in the usual Landau sense, $\alpha = \alpha(T - T_c)$, $\beta > 0$. The variation in the free energy is

$$\Delta F/V = \tfrac{1}{2}\sum_{l=1}^{\infty}(kT\alpha - \omega_l)\rho_l^2 + \ldots. \tag{4.4}$$

The temperatures at which the energy associated with non-zero mode amplitude, ρ_l, goes to zero, namely

$$T_l = (\omega_l/k\alpha)[1 - (\bar{\rho} - \rho_c)^2] \tag{4.5}$$

are the spinodal temperatures for the mode, and correspond to a curve in phase space where, in the absence of coupling between the modes, the fluctuation becomes excited spontaneously. Thus the critical fluctuations in this geometry of an isotropic lattice gas should be the predominant fluctuations at temperatures given by equation (4.5) with equations (3.10) and (3.12). The difference between this and an incoherent system, without the long-range interaction, should be demonstrable experimentally.

It is of interest to make a comparison between the present calculation for a thin, coherent, metal–hydrogen disc and that of Maier–Bötzel and Wagner [11] for a coherent metal–hydrogen system in the form of an ellipsoid, from which they take the limiting result for an oblate ellipsoid to approximate a thin disc. For the ellipsoid, the calculation is analytic and the limiting form for an oblate ellipsoid is an approximation to a thin disc, whereas the present calculation uses approximations to calculate the eigenmodes for a

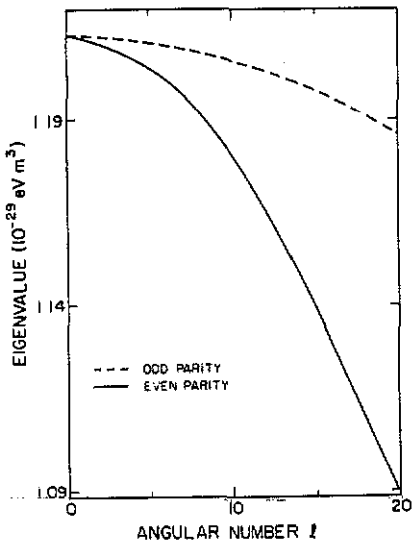

Figure 4. Eigenvalue versus l for $s = 1$, both even and odd parity modes.

cylinder, the thin, two-dimensional disc corresponding to the limit of vanishing cylinder height. The oblate ellipsoid has a continuous surface without the abrupt changes of the cylinder surfaces and the two approaches to the thin-disc problem reflect these differences in the solutions they provide. For the oblate ellipsoid [11] the eigenvalues for the limiting case of $b/a \to 0$ (b, a are the semi-axes of the ellipsoid) are, in the present notation

$$\omega_{(\text{ellipsoid})} = \mathbf{P}^2\left[\lambda + \mu\frac{(2L)(L+3)+2}{(2L)(L+3)+3}\right]^{-1} \quad \text{for } L \text{ even} \tag{4.6}$$

$$\omega_{(\text{ellipsoid})} = \mathbf{P}^2\left[\lambda + \mu\frac{2(L-1)(L+4)+6}{2(L-1)(L+4)+9}\right]^{-1} \quad \text{for } L \text{ odd} \tag{4.7}$$

with L being the eigenmode number.

For the cylinder solution, the eigenvalues for the limiting case of a disc tending to zero thickness are given by equations (4.1) and (4.2) above

$$\omega_{(\text{cylinder})} = \mathbf{P}^2[\lambda + \mu(2s+2)/(2s+3)]^{-1} \tag{4.8}$$

for both odd and even parity modes, where s is an integer (equation (3.3)). For the ellipsoid solution, with $L = 0$ and 1 in equations (4.6) and (4.7),

$$\omega_0 = \omega_1 = \mathbf{P}^2(\lambda + \tfrac{2}{3}\mu)^{-1} \tag{4.9}$$

and this eigenvalue energy corresponds to the degenerate constant hydrogen density and constant density gradient modes for which the energy may also be obtained from the cylinder solution with $l = s = 0$. The other limiting value for the surface modes,

$$\omega_\infty = \mathbf{P}^2(\lambda + \mu)^{-1} \tag{4.10}$$

is the same for both the cylinder and oblate ellipsoid solutions and these limits are consistent with the calculations of Goldberg [13]. If we compare eigenvalues given by

Table 1. Values of $\omega_{l,s}$ (with $L/R = 0.1$).

$\omega_{0,0}$	1.258	in units of
$\omega_{0,1} = \omega_{1,1}$	1.213	10^{-29} eV m^3
$\omega_{0,2} = \omega_{1,2}$	1.194	
	$\omega_{2,2} - \omega_{0,2} \approx -4 \times 10^{-4}$ even parity	
	$\omega_{2,2} - \omega_{0,2} \approx -4 \times 10^{-5}$ odd parity	

the cylinder and the oblate ellipsoid solutions for the first eigenmode of the infinitesimally thin disc for which there are coherency stresses, we get

$$\omega_{L=3}/\omega_{s=1} = (\lambda + 0.80\mu)/(\lambda + 0.92\mu) = 0.97 \tag{4.11}$$

using the values of the Lamé constants quoted in paper II. This shows the estimates to be quite close for the two methods of solution, although the respective spinodal temperatures are separated by about 15 K with T_{cyl} uppermost. For the higher eigenmodes of this disc the relationships between ω_L and ω_s are similar, with the ω_s values being higher, although both solutions have the same limit ω_∞ for the highest eigenmode.

For the higher eigenmodes obtained from the cylinder solution, where some finite thickness of the cylinder is assumed, there are two eigennumbers, s and l. The value of s effectively defines the energy and l breaks the azimuthal degeneracy. To illustrate this we use an adopted value of P (3.3 eV) and the experimental values from II: $\lambda = 1.136 \times 10^{11}$ J m^{-3}; $\mu = 0.377 \times 10^{11}$ J m^{-3} to display in table 1 the values of $\omega_{l,s}$ for a few of the higher eigenmodes.

In choosing a value of $P = 3.3$ eV for the trace of the force dipole tensor we are using past estimates as a guide but keeping in mind the observations of Pick and Bausch [19] on the consistency of estimates of values for P from different experimental data sets. Thus we use an adopted value until the matter of discrepancies in P values is more clearly resolved.

From table 1 it is clear that the change in energy involved from considering azimuthal variation is very small compared with the value of the energy, for the particular geometry chosen ($L/R = 0.1$). Effectively, the family of modes with a common s value has one characteristic energy, though there is no actual degeneracy which could give rise to 'mixing' of modes. For geometries with smaller L/R values (experimentally, ratios of 1/40 are readily attainable) inspection of equations (4.1) and (4.2) shows that the effect of the azimuthal variation on the energy eigenvalues is negligible and that such 'sheets' display eigenmodes essentially the same as those of an infinitesimally thin sheet. Differences between the cylinder and the oblate spheroid solutions, as discused above, still remain, however.

Acknowledgments

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