Surface vibrations of a ^{4}He droplet and the universality of the dispersion relation

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By applying the liquid-drop model to a ^{4}He droplet, we derive the universal dispersion relation for the droplet vibrations. Considering the eigenvalue equation for the coupled modes of surface vibrations and inner vibrations, the universality is clarified by using an effective wave number. It is shown that our universal dispersion curve for the surface modes traces well the eigenfrequencies obtained by several workers and provides benchmarks for the various methods of computer simulations. [S0163-1829(96)08621-3]

I. INTRODUCTION

The criterion for the size in which superfluidity occurs has been studied[1–4] for the finite systems of ^{4}He. As regards ^{4}He thin films many researchers have discussed the appearance condition of roton spectrum with the increase in the film thickness. For the surface modes which are called ''ripplons,'' Lauter *et al.*[3] have observed the dispersion relation against momentum transfer parallel to the surface. Gasparini, Chen, and Bhattacharyya[4] have measured the surface specific heat of confined helium. As another finite system a ^{4}He droplet has been the object for studying the dynamical structure.[5–8] Sindzingre, Klein, and Ceperley[5] have found that a ^{4}He cluster with only 64 atoms shows superfluidity.

Aiming to derive the appearance condition of superfluidity, many researchers have studied the vibrational properties of a ^{4}He droplet. Casas and Stringari[6] have calculated the vibrational frequencies by using random-phase approximation (RPA) with the density-functional formulation. Recently, Chin and Krotscheck[7] have carried out computer simulations on vibrational modes on Feynman's ansatz. For ^{4}He droplets whose number of atoms ranges from 20 to 1000, they have derived the frequencies of surface vibrations. Their calculation has been based on the variational Monte Carlo algorithm. They have plotted the dispersion relation as a function of the effective wave number $k=\sqrt{l(l+1)}/R_0$ in which R_0 is the radius of an equilibrium sphere and l is the angular momentum. Moreover, they have proposed a universal dispersion curve by choosing the k shown above. Barranco and Hernández[8] have obtained vibrational frequencies on the basis of the density-functional scheme. However, their calculated results are different from that by Chin and Krotscheck.[7]

The purpose of a present paper is to clarify the proper universality rule. We discuss the coupled modes of surface vibrations and compressional vibrations. With this end in view, we use the liquid-drop model (LDM) which has been developed to discuss dynamical structures of nuclei.[9,10]

II. LIQUID-DROP MODEL

We consider an incompressible, irrotational and nonviscous liquid. If the droplet is large, two kinds of independent vibrations occur: surface vibrations and compressional vibrations. If the droplet is small, these vibrations couple to each other.

First, to discuss the surface vibrations of an incompressible droplet, we set $R(\theta,\phi)$ to be the distance between the origin and a point on the deformed surface in the (θ,ϕ) direction. Because the angular momentum l becomes a good state variable to describe vibrations of a spherical droplet, we expand the deviation of $R(\theta,\phi)$ from an equilibrium radius R_0 by spherical harmonics $Y_{lm}(\theta,\phi)$:

$$R(\theta,\phi)=R_0+R_0\sum_l\sum_m \alpha^*_{lm}Y_{lm}(\theta,\phi), \tag{1}$$

where coefficients α^*_{lm} represent normal coordinates. From the flow velocity of the liquid drop we find the kinetic energy as

$$T=\frac{1}{2}\sum_l\sum_m B_l|\dot{\alpha}_{lm}|^2, \tag{2}$$

where $B_l=\rho_0R_0^5/l$ and ρ_0 is the mass density. Considering the excess surface area associated with the surface deformations, we obtain the potential energy to second order in α^*_{lm} as follows:

$$V=\frac{\sigma}{2}\sum_l\sum_m D_l|\alpha_{lm}|^2, \tag{3}$$

where σ represents the surface tension and $D_l=R_0^2(l-1)(l+2)$. Thus we obtain the eigenfrequencies of surface vibrations as follows:

$${}_s\omega_l=\sqrt{\sigma/(\rho_0R_0^3)}\,[l(l-1)(l+2)]^{1/2}. \tag{4}$$

In the limit of an infinite radius, we recover the dispersion relation for a planar liquid surface by the correspondence of a wave number $q=l/R_0$:

$$\omega=\sqrt{\sigma/\rho_0}\,q^{3/2}, \tag{5}$$

in which q is the wave number parallel to the surface.

Secondly, as regards compressional vibrations, we consider the wave equation for the mass density variation $\delta\rho(r,t)$:

$$\nabla^2\delta\rho(r,t)-\frac{1}{u^2}\frac{\partial^2}{\partial t^2}\delta\rho(r,t)=0, \tag{6}$$

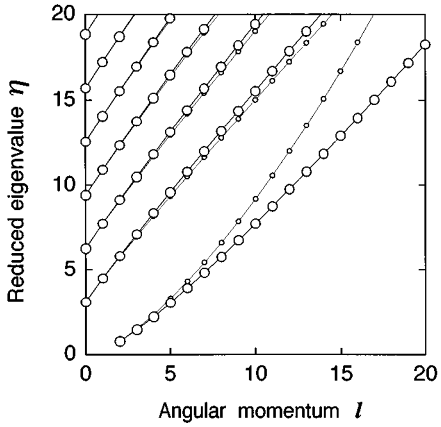

FIG. 1. Distribution of reduced eigenvalues as a function of angular momentum for a ^{4}He droplet with $N=40$. Large circles show reduced eigenfrequencies calculated by considering the coupling between surface and inner modes. Small circles show reduced eigenvalues calculated for the case without the coupling.

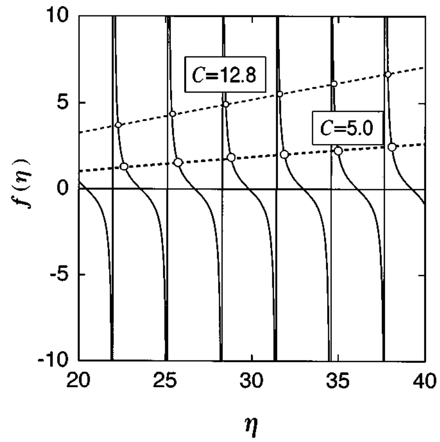

FIG. 2. Size effect on the eigenvalues for large η. Solid curves show the left-hand side of Eq. (11) and dashed curves show the right-hand side. A thin dashed-curve shows the function of the right-hand side of Eq. (11) with $C=12.8$ and a thick dashed curve shows that with $C=5.0$. Open circles represent intersections corresponding to eigenvalues.

where u is the sound velocity. The solution is written as

$$\delta\rho(r,t)=\rho_0 j_l(k_{nl}r)Y_{lm}(\theta,\phi)\alpha_{nlm}(t), \tag{7}$$

where $j_l(x)$ represents a spherical Bessel function. With the fixed boundary condition at R_0, we find the eigenvalue equation for compressional vibrations as $j_l(k_{nl}R_0)=0$ and the eigenfrequencies as ${}_c\omega_{nl}=uk_{nl}$.

Finally, we discuss the more realistic case.[10,11] If the system size is small, the surface vibrations and the compressional vibrations couple strongly to each other. In a vibrating state, therefore, the pressure caused by the surface vibrations must be in equilibrium with that caused by the compressional vibrations. By using the relation between the excess pressure $\delta p(r,t)$ associated with surface deformations and the mass density variation $\delta\rho(r,t)$: $\delta p(r,t)=u^2\delta\rho(r,t)$, we find the following eigenvalue equation for the coupled vibrations:

$$\frac{1}{j_l(\eta)}\frac{d}{d\eta}j_l(\eta)=\frac{C}{(l-1)(l+2)}\,\eta, \tag{8}$$

where C^{-1} shows the coupling constant ($C=\rho_0R_0u^2/\sigma$) and the eigenfrequencies are given as $\omega_{nl}=u\,\eta_{nl}/R_0$.

In the present LDM, the parameter C includes ρ_0, R_0, u, and σ. The change in C induces only a small change in the frequencies of the surface modes since the shift $\delta\omega$ is related to δC as follows:

$$\delta\omega=-\frac{u^2l(l-1)(l+2)}{2C^2R_0^2\omega}\,\delta C. \tag{9}$$

Because C^2 in the denominator is large (≈ 70 for $N=20$ and C^2 becomes larger as N increases), the change in u does not affect much the eigenfrequencies. Thus we set u as the sound velocity of the bulk ^{4}He liquid:[12] $u=2.37\times10^4$ (cm/s). In the same manner, the $\delta\omega$ cannot be affected by the small change in the surface tension: we use σ for a planar surface of liquid ^{4}He: $\sigma=0.354$ (dyn/cm).[13]

Figure 1 shows the distribution of reduced eigenvalues against l for the helium droplet with $N=40$ and $C=12.8$. Large circles show reduced eigenvalues calculated by Eq. (8) in which the coupling is considered. The lowest branch represents the surface modes and higher branches represent the inner modes. Small circles show reduced eigenvalues in which the coupling is neglected. Eigenvalues of the lowest branch are calculated as ${}_s\eta_l=\sqrt{l(l-1)(l+2)/C}$ and those of higher branches are calculated by $j_l(\eta)=0$.

Here we show how the coupling constant C^{-1} affects eigenvalues of noncoupled surface modes and inner modes. With the increase in C^{-1} the coupling becomes strong because a large C^{-1} means a small R_0 or a large σ. For small η we find the asymptotic eigenvalues of the surface modes by approximating $j_l(\eta)\propto\eta^l/(2l+1)!!$ where $(2l+1)!!=(2l+1)(2l-1)\cdots3\cdot1$:

$$\eta\approx{}_s\eta_l\left[1-\frac{(l-1)(l+2)}{2(2l+3)C}\right]. \tag{10}$$

This indicates that the coupling reduces the eigenvalues as C^{-1} becomes large. Moreover, the larger l, the lower the eigenvalues. For large η, we find the following eigenvalue equation for the inner modes:

$$\cot\left(\eta-\frac{\pi l}{2}\right)=\frac{C}{(l-1)(l+2)}\,\eta-\frac{l}{\eta}, \tag{11}$$

where the asymptotic form $j_l(\eta)\propto\eta^{-1}\cos\{\eta-\pi(l+1)/2\}$ is used. Figure 2 shows the size effect on the reduced eigenvalues for the mode with $l=8$. The ordinate $f(\eta)$ denotes the expression of each side of Eq. (11) and crossing points show eigenvalues. Lines normal to the η axis show the asymptotic lines given by $\eta=n'\pi$ with $7\leqslant n'\leqslant12$. In the limit $C^{-1}\to0$ (noncoupling), we find $\eta=\{n+(l/2)\}\pi$ which corresponds to the reduced eigenvalues of inner modes or bulk modes derived from $j_l(\eta)=0$. It is clear that the eigenvalues increase as the coupling constant C^{-1} increases. Thus we can conclude that the coupling between surface modes and inner modes results in the repulsion of the respective eigenvalues. The coupling becomes stronger as l increases. This is due to the situation that, for large l, the large amplitudes of the density variation $\delta\rho(r,t)$, which is expressed in terms of $j_l(kr)$, occupy the region away from the origin and approach the surface. The boundary condition at the surface, therefore, has much effect on both surface modes and inner modes.

The present LDM has been applied by Tamura and Ichinokawa[11] to explain the frequency spectrum of gallium droplets embedded in pores of a porous glass. They have assigned the observed peak[14] in the low-frequency range to the surface modes contribution and the large hump in the high-frequency range to the inner modes contribution.

To obtain the universal dispersion relation, it is necessary to estimate the size dependence of R_0. Casas and Stringari[6] have calculated R_0 as $R_0=2.22N^{1/3}$ Å (hereafter we define R_0 in units of Å) which gives the bulk density. For the calculated data by Barranco and Hernández[8] we have R_0, by the least-squares method, as $R_0=2.21N^{1/3}+0.000656+4.52N^{-1/3}$. The first term of R_0 shows the bulk contribution. The second term shows the surface contribution and the third term shows the curvature contribution. As an another example, we should note that Pandharipande *et al.*[15] have obtained the radius as $R_0=2.24N^{1/3}+0.38+2.59N^{-1/3}$ by the variational Monte Carlo method. For the calculated data by Chin and Krotscheck,[7] we have determined R_0 as $R_0=2.35N^{1/3}-0.656+5.00N^{-1/3}$, by considering their definition of the wave number $k=\sqrt{l(l+1)}/R_0$ and the sequence of their data points. Contrary to the results by Pandharipande *et al.*[15] and Barranco and Hernández,[8] the second term is negative. This is due to compensation for the large value of the first term $2.35N^{1/3}$ which underestimates the mass density of the bulk ^{4}He system in the limit $N\rightarrow\infty$.

III. UNIVERSAL DISPERSION RELATION OF VIBRATIONAL MODES

Here we derive the universal dispersion relation of the vibrational modes. To bring out the universality we introduce the mass density ρ_B and the radius R_B which satisfy

$$\rho_0 R_0^3=\rho_B R_B^3=3mN/(4\pi), \tag{12}$$

where m is the mass of a ^{4}He atom. We set $R_B=r_B N^{1/3}$ with a constant r_B which is independent of N. Consequently, ρ_B becomes independent of the droplet size; $\rho_B=3m/(4\pi r_B^3)$. We use the following R_B for the calculated data cited in Sec. II: $R_B=2.22N^{1/3}$ Å (hereafter we define R_B in units of Å) for the data by Casas and Stringari,[6] $R_B=2.21N^{1/3}$ for the data by Barranco and Hernández,[8] and $R_B=2.35N^{1/3}$ for the data by Chin and Krotscheck.[7] The R_B is the limiting radius as N tends to infinity. In contrast to ρ_B, ρ_0 depends on the droplet size because R_0 has a constant term and the term proportional to $N^{-1/3}$.

By using these parameters we transform Eq. (8) into

$$u\,\frac{\eta_{nl}}{R_e}=\sqrt{\frac{\sigma}{\rho_B}}\left[\frac{R_e^3}{l(l-1)(l+2)}+\frac{\sigma R_e^2}{\rho_B u^2}\,\frac{j_{l+1}(\eta_{nl})}{l\,\eta_{nl} j_l(\eta_{nl})}\right]^{-1/2}, \tag{13}$$

where we introduce the effective radius $R_e=R_B^3/R_0^2$ and use the relation

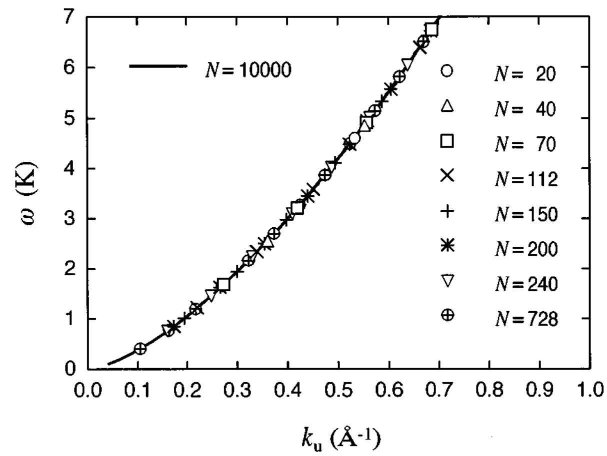

FIG. 3. Dispersion relation of surface modes as a function of k_u. Vibrational frequencies are shown in units of Kelvin. All symbols show the eigenfrequencies of the surface modes with R_0 and R_B determined from the data by Barranco and Hernández (Ref. 8). Calculated data are on a universal curve irrespective of droplet sizes.

$$\frac{dj_l(\eta)}{d\eta}=\frac{l}{\eta}\,j_l(\eta)-j_{l+1}(\eta). \tag{14}$$

It is essential that the coefficient $u\sqrt{\rho_B/\sigma}$ is independent of the droplet size. This is a reason why we introduce ρ_B and R_B to obtain the universal dispersion relation. From Eq. (13) we define the effective wave number k_u as

$$k_u=\left[\frac{R_e^3}{l(l-1)(l+2)}+\frac{\sigma R_e^2}{\rho_B u^2}\,\frac{j_{l+1}(\eta_{nl})}{l\,\eta_{nl} j_l(\eta_{nl})}\right]^{-1/3}$$
$$=\left[\frac{\sigma R_e^2}{\rho_B u^2\eta_{nl}^2}+\left(1-\frac{\rho_0 R_0^3}{\rho_B R_B^3}\right)\frac{R_e^3}{l(l-1)(l+2)}\right]^{-1/3}, \tag{15}$$

in which we use

$$\frac{j_{l+1}(\eta_{nl})}{j_l(\eta_{nl})}=\frac{1}{\eta_{nl}}-\frac{\rho_0 R_0 u^2}{\sigma}\,\frac{1}{(l-1)(l+2)}\,\eta_{nl}. \tag{16}$$

Because $\rho_0 R_0^3=\rho_B R_B^3$, the second term in round brackets of Eq. (15) is zero. This is an another reason why we introduce ρ_B and R_B. Thus we obtain the simpler expression for k_u as

$$k_u=\left(\frac{\rho_B}{\sigma}\right)^{1/3}\left(u\,\frac{\eta_{nl}}{R_e}\right)^{2/3}. \tag{17}$$

We should note that k_u is a discrete variable. With the definition $\omega_u=u\,\eta_{nl}/R_e$, we obtain the universal dispersion relation as follows:

$$\omega_u=\sqrt{\sigma/\rho_B}\,k_u^{3/2}. \tag{18}$$

Except the definition of the k_u associated with the effective radius R_e, this is of the same type as that of a planar liquid surface.

IV. CALCULATED RESULTS AND DISCUSSION

For the data by Barranco and Hernández,[8] we show the universal dispersion relation against k_u in Fig. 3. All calcu-

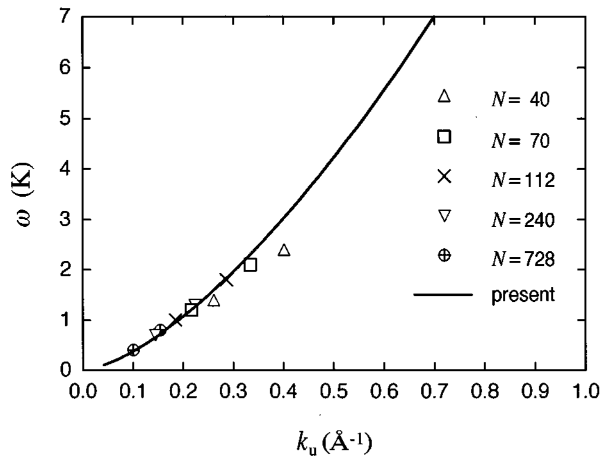

FIG. 4. Dispersion relation of surface modes as a function of k_u. Vibrational frequencies are shown in units of Kelvin. All symbols show the eigenfrequencies of the surface modes with R_0 and R_B determined from the data by Casas and Stringari (Ref. 6): $R_0=2.22N^{1/3}$ and $R_B=2.22N^{1/3}$. Eigenfrequencies for different sizes are on a universal curve.

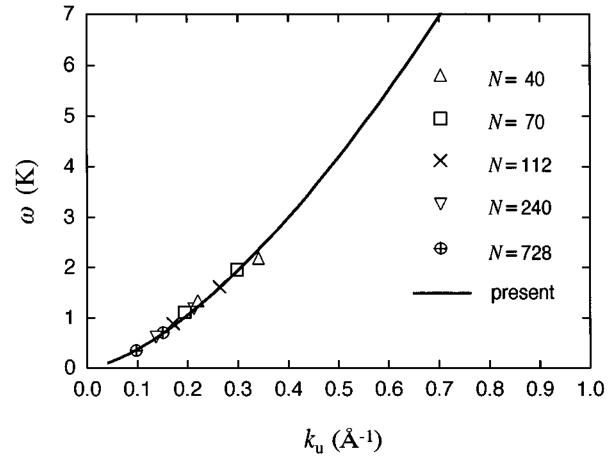

FIG. 5. Dispersion relation of surface modes as a function of k_u. Vibrational frequencies are shown in units of Kelvin. All symbols show the eigenfrequencies of the surface modes with R_0 and R_B determined from the data by Barranco and Hernández (Ref. 8): $R_0=2.21N^{1/3}+0.000656+4.52N^{-1/3}$ and $R_B=2.21N^{1/3}$. Eigenfrequencies for different sizes are on a universal curve.

lated data are on the universal dispersion curve irrespective of droplet sizes. In our treatment, we assume that a ^{4}He droplet is the continuum system. Actually, the droplet consists of atoms. To incorporate the discreteness into the present continuum system, we set the maximum angular momentum. This means that the vibrational modes whose wavelengths are less than twice the mean interatomic distance cannot exist. In Refs. 11 and 16, this maximum is defined as

$$l_{\max}=\left[\frac{\pi}{d}\,R_e-\frac{1}{2}\right], \tag{19}$$

where $[x]=j$ for $j\leqslant x<j+1$ and d represents the mean interatomic distance. For the data by Barranco and Hernández[8] we have $l_{\max}=5$, 7, 8, 11, and 16 for $N=40$, 70, 112, 240, and 728, respectively. With this maximum angular momentum, we have the proper frequency spectrum of the droplet.[11] If we use a simple Debye model in which only the maximum of frequencies is introduced, we have an improper frequency spectrum. The $l_{\max}$ corresponds to the Brillouin-zone boundary of the bulk system having the translational symmetry. For the detail we refer to Refs. 11 and 16.

Figure 4 shows calculated frequencies by Casas and Stringari[6] and ours as a function of k_u specified by their R_0 and R_B. Our universal curve agrees well with their calculated data for droplets with $N\geqslant 100$ and in the range $k_u\leqslant 0.3$ Å^{-1}. Figure 5 shows calculated frequencies by Barranco and Hernández[8] and our universal curve. Similarly to Fig. 4, our universal curve agrees with their calculated data for droplets with $N\geqslant 70$ and in the range $k_u\leqslant 0.3$ Å^{-1}. Evidently, our universal curves in both Figs. 4 and 5 trace their data points though methods of calculations by Casas and Stringari[6] and Barranco and Hernández[8] are different to each other. Moreover, even if N is small, the eigenvalues with small l can be described well by the present LDM.

Figure 6 shows calculated frequencies by Chin and Krotscheck[7] and our universal curve. Our universal curve is only in qualitative agreement with their data. Even if we use R_0 by Barranco and Hernández,[8] we cannot explain the sequence of their data. This is in contrast to the result that our dispersion curve traces well the calculated results by Casas and Stringari[6] and Barranco and Hernández.[8] Thus there remains a serious problem because in the low wave-number range the dispersion curve must be described by LDM. Concerning the eigenfrequencies of surface vibrations, we support the results calculated by Casas and Stringari[6] and by Barranco and Hernández.[8]

In the range $k_u>0.3$ Å^{-1}, we cannot explain the lowering of vibrational frequencies. This is due to the assumption that the droplet has an abrupt surface defined by Eq. (1). Casas and Stringari[6] have studied the effect of surface diffuseness on the vibrational frequencies. They have found that the low-

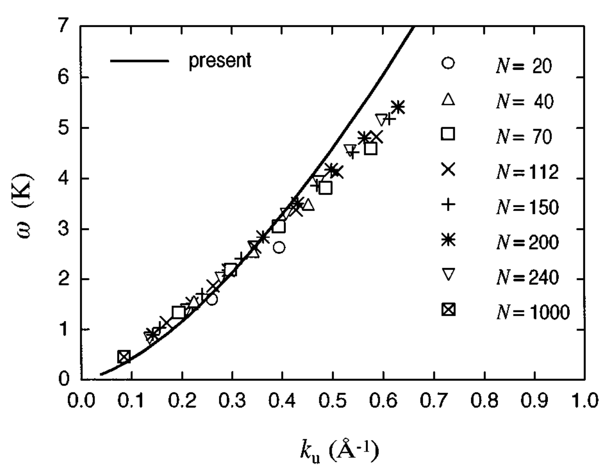

FIG. 6. Dispersion relation of surface modes as a function of k_u. Vibrational frequencies are shown in units of Kelvin. All symbols show the eigenfrequencies of the surface modes with R_0 and R_B determined from the data by Chin and Krotscheck (Ref. 7): $R_0=2.35N^{1/3}-0.656+5.00N^{-1/3}$ and $R_B=2.35N^{1/3}$. Our dispersion curve is in poor correspondence with their calculated data.

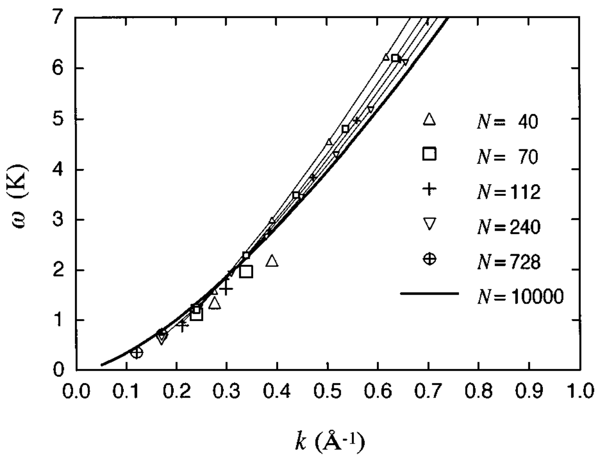

FIG. 7. Dispersion relation of surface modes as a function of k introduced by Chin and Krotscheck (Ref. 7). Vibrational frequencies are shown in units of Kelvin. Eigenfrequencies shown by large symbols are those calculated by R_0 determined from the data by Barranco and Hernández (Ref. 8). Small symbols show our dispersion relations calculated by LDM. There is no universality in the dispersion relation.

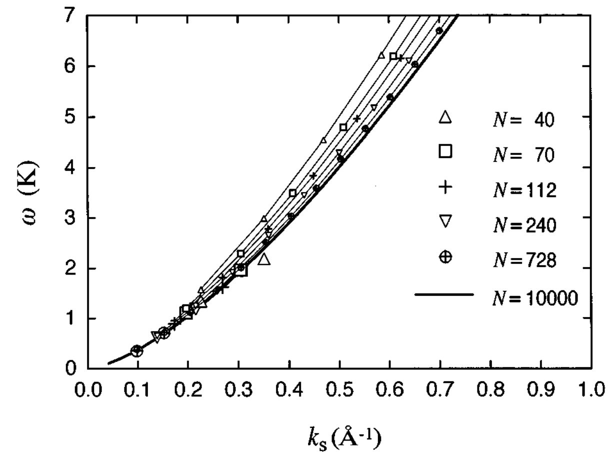

FIG. 8. Dispersion relation of surface modes as a function of k_s. Vibrational frequencies are shown in units of Kelvin. Eigenfrequencies shown by large symbols are those calculated with R_0 determined from the data by Barranco and Hernández (Ref. 8). Small symbols show our dispersion relations calculated by LDM. There is no universality in the dispersion relation.

ering of eigenfrequencies of the surface vibrations occurs with the increase in the surface diffuseness and the critical number of atoms is about 100. In accordance with this criterion, Fig. 4 shows that our dispersion curve deviates from their data for a ^{4}He droplet with 40 atoms. By taking the distribution of the mass density to be of a type of Fermi-Dirac distribution function, we can consider, within the framework of the present LDM, the effect of the surface diffuseness on the dispersion relation. Expanding the mass density around a certain radius, we obtain the kinetic energy and the potential energy with corrections coming from the surface diffuseness. This procedure is carried out in the same way as deriving the heat capacity of a free-electron system at finite temperatures. We find that the eigenfrequencies reduce with the decrease in N and the increase in l. These results will be reported elsewhere. If a ^{4}He droplet shows superfluidity, it is thought that the structure factor $S(k)$ introduced by Feynman[17] lowers the dispersion curve more.

In Fig. 7 we show the dispersion relation by using an effective wave number $k=\sqrt{l(l+1)}/R_0$ proposed by Chin and Krotscheck.[7] Obviously, there is no universality in the dispersion curve because eigenfrequencies derived by LDM are scattered. The k has been introduced by considering the conservation of momentum associated with the passing probe. We, however, cannot specify the feature of the droplet because such a k must be common to a liquid drop and a particle in a solid state. Even if we use $k_s=[l(l-1)(1+2)]^{1/3}/R_0$, which has been suggested by Chin and Krotscheck,[7] there still remains the scattering of eigenfrequencies especially in the range of high wave numbers as is shown in Fig. 8. In our formulation we obtain, for small η_{nl}, the wave number associated with the surface modes as

$$k_u \approx [l(l-1)(1+2)]^{1/3}/R_e, \tag{20}$$

in which the asymptotic relation $\eta^2=\sigma l(l-1)(l+2)/(\rho_0 R_0 u^2)$ is used in Eq. (17). This indicates that the denominator R_e improves a simple definition of a wave number $k_s=[l(l-1)(1+2)]^{1/3}/R_0$ derived from Eq. (4). We should note that the difference between R_0 and R_e increases with the decrease in N and the k_s gives many deviations from the universal dispersion relation. This is the reason for deviations from the universal dispersion curve.

V. CONCLUSIONS

We have clarified the vibrational properties of a ^{4}He droplet by the liquid-drop model which includes the coupling between surface vibrations and inner vibrations. For different sizes of ^{4}He droplets, we have shown that the universal dispersion relation can be specified by introducing the effective wave number k_u. Because our method gives a proper universal curve, our dispersion relation serves as a test of the calculated results on various kinds of schemes such as variational Monte Carlo, diffusion Monte Carlo, RPA and so forth.

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