Lagrangian formulism of elasticity with relevance to surface energy

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Abstract

By introducing the divergence of a vector potential into the Lagrangian, a Lagrangian framework is developed to incorporate the surface energy into elasticity. Besides the Euler-Lagrange equation and natural boundary condition, a new boundary constitutive equation is derived from the variation of the Lagrangian and configuration on which the Lagrangian is defined. On the boundary surface, explicit expression of the vector potential with respect to field variable and surface curvature is determined. Based on this framework, an elastic model with relevance to the surface energy is established. The Young-Laplace’s formula is generalized into elastic solid in a new form. Making use of this model, we investigate the surface energy effect in the radial vibration of spherical nanoparticle. Numerical calculation shows that natural frequencies of nanoparticle will shift down due to the surface energy. This shift is especially apparent in the vibration of soft matter nanoparticles.

Key words: Euler-Lagrange equation, surface energy, generalize Young-Laplace’ formula, natural frequency, nanoparticle

1 Introduction

From the view of atomic level, the free surface of a bulk material is different from its interior. This difference gives rise to a excess energy to appear on surface. It is called the surface free energy (or shortly called the surface energy). In classical continuum mechanics, the influence of surface energy on the deformation of solid is usually ignored, because atoms on surface is far smaller in magnitude than that in the interior of bulk. However, with entering from microscale into nanoscale, the ratio of atoms on surface to total atoms becomes gradually significant. Therefore, the surface energy effect has to be taken into account in the deformation of solid.

On the surface energy effect in elastic deformation, early works can be traced back to Shuttleworth [1], Herring [2], Nicolson [3] and Vermaak et al [4]. They investigated the lattice contraction caused by the surface tension in small crystal particles. A general theoretical framework incorporating surface energy into continuum mechanics was proposed in the 1970s by Gurtin and Murdoch [5, 6]. Within this framework, Steigmann and Ogden [7] generalized the conception of surface energy to the curvature dependence case. Huang and Wang et al [8, 9] clarified the necessity to introduce three configurations in an accurate and complete description for the deformation of continuum with surface energy. Altenbach et al [10] gave a proof on existence and uniqueness of the solutions of initial and boundary value problems of linear elasticity with surface energy. Schiavone and Ru [11] discussed the solvability of boundary value problems in a theory of plane-strain elasticity concerned with surface energy.

Various models based on the Gurtin-Murdoch theory have been presented in several contexts, e.g. nanoparticles, wires and films (Huang and Thomson et al [12]; Cammarata [12, 13]; Dingreville and Qu et al [15]; Park [16]), nanoscale rods, beams, plates and shells (Miller and Shenoy [17]; Huang [18]; Bar and Altus et al [19]; Liu and Rajapakse [20]; Wang and Feng [21]; Wang and Zhao et al [22]), nano inclusions (Duan and Wang et al [23]; He and Li [24]; Sharma and Ganti [25]). So far, a considerable number of literatures have been devoted to the continuum models considering surface energy. It is not the purpose of this paper to list and review these abundant literatures. The reader can be referred to the recent reviews by Wang and Huang et al [26] and Duan, Wang and Karihaloo [27] on the literatures.
The subject of the paper will focus on developing a Lagrangian formulation corresponding to the Gurtin-Murdoch theory. The novel feature of this formulation consists in that a vector potential is introduced to characterize the surface energy of continuum. Consequently, the Euler-Lagrange equation, natural boundary condition and a new boundary constitutive equation can be consistently determined by simultaneously taking the variation of the Lagrangian and configuration of continuum on which the Lagrangian is defined.

The paper is outlined as follows. In section 2, based on a vector potential, a Lagrangian framework is developed to incorporate surface energy into continuum mechanics. The boundary constitutive equation and the vector potential are determined on the boundary surface. By linearization, in section 3 we put forward to an elastic model with relevance to surface energy in which a generalized Young-Laplace’s formula is derived consistently. Making use of this elastic model, we investigate the surface energy effect in the radial vibration of spherical nanoparticle. Some interesting results are predicted. Finally, we close this paper with a summary.

Notation: A compact notation is used, with boldface letters being vectors or tensors. The index rules and summation convention are adopted. Latin indices have the range 1, 2, 3. Partial derivatives with respect to coordinates are represented as \( \partial_k = \partial/\partial x^k \) or \( (\cdot)_k = \partial(\cdot)/\partial x^k \). Partial derivative with respect to time is denoted by an upper dot, e.g., \( \dot{a} = \partial a/\partial t \). Other symbols will be introduced in the text where they appear for the first time.

2 Theoretical framework

Let \( x = \{x^j\} \) \((j = 1, 2, 3)\) be a 3-dimensional position vector on \( \Omega \subset \mathbb{R}^3 \). The Greece letter \( \Omega \) stands for current configuration of a continuum. We use \( \varphi = \varphi(t, x) \) to characterize the field defined on \([t_0, t_1] \cup \Omega\), where \( t \in [t_0, t_1] \) denotes time. Depending on circumstances, \( \varphi \) is a scalar, vector or tensor.

As is well known, if two Lagrangians differ by the divergence of a vector function only of the field variables and coordinates, their Euler-Lagrange equations are the same in form. Thus, if such a divergence term is added to a Lagrangian, dynamic behaviors of field are unchanged under the fixed boundary condition. However, if the boundary is free, the additive divergence will contribute to the field by entering the variation of the Lagrangian but also the variation of the surface area and volume of continuum. Under circumstances taking the variation of the Lagrangian and configuration of continuum on which the Lagrangian is defined. Consequently, the calculation for the variation of \( A[\varphi] \) should not only include the variation of the Lagrangian but also the variation of the surface area and volume of continuum. Under

$$A[\varphi] = \int_{t_0}^{t_1} \int_{\Omega} [L(\varphi, \varphi, \partial_j \varphi) - \partial_k Y^k(x, \varphi)] dv(x) dt.$$  

(1)

In terms of the divergence theorem, \( A[\varphi] \) can be also written as

$$A[\varphi] = \int_{t_0}^{t_1} \int_{\Omega} L(\varphi, \dot{\varphi}, \partial_j \varphi) dv(x) dt - \int_{t_0}^{t_1} \int_{\partial \Omega} Y^k(x, \varphi) n_k da(x) dt,$$  

(2)

where \( n_k \) denotes the unit normal vector on the boundary \( \partial \Omega \) of \( \Omega \). It is necessary to emphasize the distinction between the vector potential and surface energy. The former is defined on continuum, including its interior and surface; but the latter is only on the surface. In the following, we will demonstrate that they are equivalent on surface.

If surface energy is concerned, the changes of the configuration of continuum will become a factor having to be considered. Consequently, the calculation for the variation of \( A[\varphi] \) should not only include the variation of the Lagrangian but also the variation of the surface area and volume of continuum. Under
this consideration, the variation of $A[\varphi]$ can be calculated as follows:

$$
\delta A[\varphi] = \int_{t_0}^{t_1} \int_{\Omega} \delta L dv(x) dt + \int_{t_0}^{t_1} \int_{\partial \Omega} L dv(x) dt \\
- \int_{t_0}^{t_1} \int_{\partial \Omega} \delta Y^k n_k da(x) dt - \int_{t_0}^{t_1} \int_{\delta(\partial \Omega)} Y^k n_k da(x) dt
$$

$$
= \int_{t_0}^{t_1} \int_{\Omega} \left[ \frac{\partial L}{\partial \varphi} \delta \varphi + \frac{\partial L}{\partial Y^j} \delta \varphi + \frac{\partial L}{\partial \varphi_j} \delta \varphi \right] dv(x) dt \\
- \int_{t_0}^{t_1} \int_{\partial \Omega} \frac{\partial Y^k}{\partial \varphi} n_k \delta \varphi da(x) dt + \int_{t_0}^{t_1} \int_{\partial \Omega} L dv(x) dt - \int_{t_0}^{t_1} \int_{\delta(\partial \Omega)} Y^k n_k da(x) dt
$$

$$
= \int_{t_0}^{t_1} \int_{\Omega} \left[ \frac{\partial L}{\partial \varphi} \frac{d}{dt} \left( \frac{\partial L}{\partial \varphi} \right) - \partial_j \left( \frac{\partial L}{\partial \varphi_j} \right) \right] \delta \varphi dv(x) dt + \int_{t_0}^{t_1} \int_{\partial \Omega} \frac{\partial L}{\partial \varphi} n_k \delta \varphi da(x) dt \\
+ \int_{t_0}^{t_1} \int_{\partial \Omega} L dv(x) dt - \int_{t_0}^{t_1} \int_{\delta(\partial \Omega)} Y^k n_k da(x) dt,
$$

in which we have supposed that $\varphi(x)$, $L(\varphi, \varphi, \varphi_j)$ and $Y^j(x, \varphi)$ are suitably smooth functions. On-Yang and Helfrich [28] have proved that

$$
\int_{\delta \Omega} \delta Y^k n_k da(x) = \int_{\Omega} 2H g_{kj} Y^k \delta u^j da(x),
$$

where $\delta u^j$ is the variation of displacement of the area element $da(x)$ on the boundary surface $\partial \Omega$, $H$ the mean curvature at $x$ on $\partial \Omega$ and $g_{kj}$ the materic tensor. $\delta \varphi = 0$ at the two end points of time, so substituting Eq. (4) and (5) into (3) yields

$$
\delta A[\varphi] = \int_{t_0}^{t_1} \left[ \frac{\partial L}{\partial \varphi} \frac{d}{dt} \left( \frac{\partial L}{\partial \varphi} \right) - \partial_j \left( \frac{\partial L}{\partial \varphi_j} \right) \right] \delta \varphi dv(x) dt + \int_{t_0}^{t_1} \int_{\partial \Omega} \frac{\partial L}{\partial \varphi} n_k \delta \varphi da(x) dt \\
+ \int_{t_0}^{t_1} \int_{\partial \Omega} (L n_j - 2H g_{kj} Y^k) \delta u^j da(x) dt.
$$

According to the Hamilton’s principle, $\delta A[\varphi] = 0$. Therefore, the fundamental lemma of variation leads to the below results:

Euler-Lagrange equation:

$$
\frac{d}{dt} \left( \frac{\partial L}{\partial \varphi} \right) + \partial_k \left[ \frac{\partial L}{\partial \varphi_k} \right] - \frac{\partial L}{\partial \varphi} = 0.
$$

Natural boundary condition:

$$
\left[ \frac{\partial L}{\partial \varphi_j} \right]_{\partial \Omega} = 0.
$$

Boundary constitutive equation :

$$
(L n_j - 2H g_{kj} Y^k)_{\partial \Omega} = 0.
$$

Eq. (7) and (8) show that a divergence term, only relevant to the field variable, added to the Lagrangian has no effects on the Euler-Lagrange equation, but it contributes to the natural boundary condition of field. Since $Y^k$ depends only on $\varphi$, $H$ in Eq. (9) is necessarily relevant to $\delta \varphi$. Or else the derivative of $L$ with respect to $\delta \varphi$ will be zero, in terms of Eq. (9). That contradicts with Eq. (8). Making use of Eq. (9), we have

$$
\left[ \frac{\partial L}{\partial \varphi_j} n_j - 2g_{kj} Y^k \frac{\partial H}{\partial \varphi_j} \right]_{\partial \Omega} = 0.
$$

Comparing Eq. (10) with (8), one can see that $H$ is linearly dependent of $\delta \varphi$. Subtracting Eq. (9) from (10) gives

$$
\left[ \frac{\partial Y^k}{\partial \varphi} n_k - 2g_{kj} Y^k \frac{\partial H}{\partial \varphi_j} \right]_{\partial \Omega} = 0.
$$
Let \( n^j \) denote the contravariant component of the unit normal vector on \( \partial \Omega \). So \( n^j n_k = \delta^j_k \). By this identity, Eq. (11) can be rewritten as
\[
\left. \frac{\partial \Gamma}{\partial \varphi} - S \right|_{\partial \Omega} = 0,
\]
where
\[
\Gamma = Y^k n_k, \quad S = 2 n_k \frac{\partial H}{\partial (\partial k \varphi)}.
\]
On \( \partial \Omega \), the solution of Eq. (12) can be represented as
\[
\Gamma = \gamma \exp(S \varphi),
\]
where \( \gamma \) is an integral constant. Inserting Eq. (13) in (14) leads to
\[
Y^k n_k = \gamma \exp[2 \varphi n_k \frac{\partial H}{\partial (\partial k \varphi)}],
\]
which characterizes the mathematical form of \( Y^k n_k \) on the free surface of \( \Omega \). Eq. (15) shows that, on surface, \( Y^k n_k \) (i.e., \( \Gamma \)) is a surface curvature-dependent energy. So it is just the surface energy. It should be noticed that, due to concerning the curvature of surface, Eq. (15) holds only on the surface \( \partial \Omega \), but not valid in the interior of \( \Omega \).

3 Linear elastic model with relevance to surface energy

Consider a free elastic body concerned with surface energy. The Lagrangian of it is assumed to take the following form (In the following, all indices are written as subscripts without distinguishing the superscripts and subscripts.):
\[
L = L - Y_{k,k} = \frac{1}{2} \rho \ddot{u}_k \dot{u}_k - \frac{1}{2} C_{ijkl} u_{i,j} u_{k,l} - Y_{k,k}.
\]
where \( u_k \) denotes the elastic displacement field, \( \rho \) and \( C_{ijkl} \) are the mass density and elastic tensor. Based on the viewpoint of Huang and Wang [8], \( Y_{k,k} \) can be regarded as elastic energy transformed from surface energy relative to the so-called fictitious stress-free configuration. In terms of Eq. (14), \( Y_k n_k \) is given by
\[
Y_k n_k = \Gamma = \gamma \exp(S_k u_k).
\]
Under the small displacement condition, Eq. (17) is expanded to the second order term of \( u_k \), i.e.,
\[
Y_k n_k = \gamma (1 + S_k u_k + \frac{1}{2} S_k S_j u_k u_j),
\]
where \( S_k \) is determined by Eq. (13)\(_2\). Just mentioned above, the mean curvature \( H \) in Eq. (13)\(_2\) is linearly dependent of \( u_k,i \). As thus, \( H \) is only represented as \( H = H_0(1 + u_k,k) \) in terms of the invariance of \( H \) [29]. Here, \( H_0 \) is the initial mean curvature. Making use of Eq. (13)\(_2\), we have \( S_k = 2 n_k H_0 \). With help of this result, Eq. (18) becomes
\[
Y_k n_k = \gamma (1 + 2 H_0 u_k n_k + 2 H_0^2 u_k n_k u_j n_j).
\]
By means of Eq. (16) and (19), Eq. (7) and (8) can be written as
\[
C_{ijkl} u_k,j = \hat{\rho} \ddot{u}_i, \quad \text{in } \Omega.
\]
\[
C_{ijkl} u_k,j n_j - 2 \gamma H_0 n_i (1 + 2 H_0 u_k n_k) = 0, \quad \text{on } \partial \Omega.
\]
Eq. (20) and (21) are the equation of motion and boundary condition to characterize the free elastic body concerned with surface tension, respectively. By means of the Hooke’s law \( \sigma_{ij} = C_{ijkl} u_k,l,i \), Eq. (21) is rewritten as
\[
\sigma_{ij} n_j = 2 \gamma H_0 n_i (1 + 2 H_0 u_k n_k).
\]
If the term \( 2 H_0 u_k n_k \) is dropped, Eq. (22) will reduce to the Young-Laplace’s formula that describes the surface pressure of fluid. Therefore, Eq. (22) can be regarded as a generalization of the Young-Laplace’s formula in solid, whereas \( \gamma \) represents the specific surface energy factor.
4 Application to the radial vibration of nanoparticle

4.1 Characteristic frequency equation

As an example, we consider the radial free vibration of a spherical nanoparticle with the radius of \( r_0 \). Let material be isotropic. The equations governing this vibration can be determined by Eq. (20) and (21) together with \( H_0 = -1/r_0 \) and \( C_{ijkl} = \lambda \delta_{ij} \delta_{kl} + \mu (\delta_{ik} \delta_{lj} + \delta_{il} \delta_{jk}) \). In a spherical coordinate system, they are written as

\[
\frac{\partial^2 u_r}{\partial r^2} + \frac{2 \partial u_r}{r \partial r} - \frac{u_r}{r^2} = \frac{1}{c^2} \frac{\partial^2 u_r}{\partial \tau^2},
\]

\[
[(\lambda + 2\mu)\frac{\partial u_r}{\partial r} + 2(\lambda - 2\gamma)u_r]_{r=r_0} = -\frac{2\gamma}{r_0},
\]

where \( \lambda \) is the Lamé constant and \( \mu \) the shear modulus. \( c = \sqrt{ (\lambda + 2\mu)/\rho} \) being the wave velocity. Assume that the solution of Eq. (23) subjected to (24) to have the form below:

\[
u_r(r, \tau) = \bar{u}_r + g(r)e^{i\omega \tau},
\]

where \( \bar{u}_r \) reads

\[
\bar{u}_r = -\frac{2\gamma}{(3\lambda + 2\mu)r_0 - 4\gamma r},
\]

which is the radial contraction displacement caused by the surface energy \([12]\). It should be noted that \( \bar{u}_r > 0 \) or \( \bar{u}_r \to \infty \) when \( r_0 \leq r_c = 4\gamma/(3\lambda + 2\mu) \). That is impossible in physics because it indicates that the surface energy will cause the radial extension of a spherical particle. Therefore, the elastic model is no longer valid for the case that \( r_0 \leq r_c \). Inserting Eq. (25) in (23) and (24) leads to

\[
\frac{\partial^2 g}{\partial r^2} + \frac{2 \partial g}{r \partial r} + (\frac{\omega^2}{c^2} - \frac{2}{r^2})g = 0.
\]

\[
[(\lambda + 2\mu)\frac{\partial g}{\partial r} + 2(\lambda - 2\gamma)\frac{\partial g}{r}]_{r=r_0} = 0.
\]

Let \( k = \omega/c \) and \( g(r) = f(r)/\sqrt{r} \). By means of these substitution, Eq. (27) can be transformed into a standard Bessel equation. Therefore, the general solution to Eq. (28) can be given as follows:

\[
g(r) = Ar^{-1/2}J_{3/2}(kr) + Br^{-1/2}Y_{3/2}(kr),
\]

where \( J_{3/2}(kr) \) and \( Y_{3/2}(kr) \) are first kind and second kind Bessel function of order \( 3/2 \), respectively. \( A \) and \( B \) are two undetermined constants. Noticing \( g(0) = 0 \), we have \( B = 0 \). Thus, Eq. (29) reduces to

\[
g(r) = Ar^{-1/2}J_{3/2}(kr).
\]

Substituting Eq. (30) into (28) yields

\[
k J'_{3/2}(kr_0) + \frac{(3\lambda - 2\mu)r_0 - 8\gamma}{2(\lambda + 2\mu)r_0^2} J_{3/2}(kr_0) = 0,
\]

where \( J'_{3/2} \) denotes the derivative of \( J_{3/2} \) with respect to its argument. Eq. (31) represents the characteristic frequency equation of the radial vibration of the spherical nanoparticle involving the surface energy effect.

4.2 Results and discussion

Using Eq. (31), we calculate the natural frequencies of two different nanoparticles. The first is the silica gel particle, and the second is the palladium (Pd) particle. The silica gel is a soft matter. We take its \( E = 2.14 \text{MPa}, \nu = 0.48, \rho = 1200 \text{kg/m}^3 \) and \( \gamma = 22.0 \text{mN/m} \). In terms of the transformation \( \lambda = E\nu /[(1 + \nu)(1 - 2\nu)] \) and \( \mu = E/[(1 + 2\nu)] \), we have \( \lambda = 17.35 \text{MPa} \) and \( \mu = 0.72 \text{MPa} \). As thus, \( r_c = 4\gamma/(3\lambda + 2\mu) = 1.65 \text{mm} \). When calculating, the range of \( r_0 \) should be greater than 1.65nm.

The changes of first three natural frequencies with the radius \( r_0 \) are shown in Figure 1. It can be seen that the three natural frequencies increase with a decrease in the radius. This is a well known size effect. More significantly, Figure 1 illustrates that, with entering the nano-level, the influence of surface energy on the natural frequency becomes apparent gradually. The natural frequencies will shift down due to the surface energy. For the first frequency, the shifting magnitude comes up to 33.5% of the original value at
Figure 1: The changes of first three natural frequencies with the radius of the silica gel nanoparticle

$r_0 = 2.5$nm, and 12.8% at $r_0 = 5$nm. Even when $r_0 = 50$nm, this ratio can also reach 1.2%. Therefore, the surface energy effect is not negligible in the vibration of the silica gel nanoparticle.

On the other hand, the influence of surface energy on the higher order frequency is far smaller than that on the first frequency. At $r_0 = 2.5$nm, the calculation shows that the shift of third frequency caused by the surface energy is less than 2% of this frequency. Therefore, for the more higher frequency, the surface energy effect can be ignored.

Figure 2: The changes of the first natural frequencies with the radius of the Pd nanoparticle

Taking $E = 123.6$GPa, $\nu = 0.39$, $\rho = 12023$kg/m$^3$ and $\gamma = 6.0$N/m [30], we investigate the radial vibration of the Pd particle with the surface energy effect. The results are shown in Figure 2, which depicts the first natural frequency of the Pd particle changing with radius. From it, it can be seen that the influence of surface energy on the first natural frequency is extremely slight. The calculation shows that the frequency shifting caused by the surface energy is about 3% of this frequency when $r_0 = 0.5$nm. Therefore, the surface energy effect can be almost ignored in the vibration of the Pd nanoparticle.
The results above indicate that, in the vibration, the influence of surface energy on the silica gel nanoparticle differs from that on the Pd nanoparticle. From the view of theory, this difference is determined by relative ratio the elastic modulus to the surface energy factor. Palladium is a "hard" material. Its surface energy factor $\gamma$ is far smaller than the elastic modulus in magnitude. This makes that the influence of $\gamma$ on Eq. (31) is extremely slight so that it can be ignored. However, silica gel is a soft matter. At the nano-level, the ratio $\gamma/\gamma_0 > 1$. Therefore, the surface energy effect must be considered in the vibration of the silica gel nanoparticle.

5 Summary

A vector potential depending only on the field variables and coordinates is defined. By introducing the divergence of this vector potential into the Lagrangian, a Lagrangian framework is developed to incorporate surface energy into the theory of elasticity. Besides the Euler-Lagrange equation and natural boundary condition, a new boundary constitutive equation is derived from the variation of surface area and volume of continuum. The boundary constitutive equation characterizes the relation, on the boundary surface of continuum, between the Lagrangian, the vector potential and the mean curvature of surface. Explicit expression of the vector potential with respect to the field variable and the mean curvature is also determined on the boundary surface.

Based on this framework, an elastic model with relevance to surface energy is established. In this model, a generalized Young-Laplace’s formula is derived consistently. Compared with the existing results, the generalized Young-Laplace’s formula depends not only on surface energy and surface curvature but also on the normal displacement of surface. Making use of this model, we investigate the surface energy effect in the radial vibration of spherical nanoparticle. By calculation, some conclusions are given as follows:

(1) At the nano-level, the influence of surface energy on the natural frequencies of a solid particle depends on the relative difference between elastic modulus and the surface energy factor of material in magnitude. The surface energy effect can be ignored in the vibration of hard nanoparticles, but for soft nanoparticles, it is necessary to take this effect into account.

(2) For a soft nanoparticle, its natural frequencies will shift down due to the surface energy effect. The smaller the size is, the stronger the frequencies shifting.

(3) In the vibration of soft nanoparticle, the influence of surface energy on first natural frequency is far greater than that on the other frequencies. For the higher order frequencies beyond the third frequency, the surface energy effect can be ignored.

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