https://doi.org/10.15407/ujpe63.2.150

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WETTING UNDER ELECTROMAGNETIC RESONANCE IRRADIATION

The influence of the resonance electromagnetic irradiation on the wetting of a solid surface by liquid has been discussed. A simple model of a fluid consisting of two-level atoms, for which changes in their interaction due to a resonance irradiation can be found in the framework of the quantum-mechanical perturbation theory is considered, and the corresponding functional for the grand thermodynamic potential is found. The density functional method is used to calculate the surface tension at the liquid–vapor, solid–liquid, and solid–vapor interfaces, and the Young equation is applied to determine the wetting angle. It is shown that the resonance irradiation can significantly increase the latter parameter.

Keywords: resonance irradiation, density functional method, surface tension, wetting angle.

1. Introduction

The study of the solid surface wetting by a liquid is important from both the theoretical and practical viewpoints [1–4]. They say that a liquid wets a solid surface, if the Young equilibrium wetting angle θ, i.e. the angle between the liquid and solid surfaces under thermodynamic equilibrium conditions, tends to zero, and that it does not do it if θ tends to 180°. The wetting angle can change, if external parameters (e.g., the temperature) are varied. The density functional method [5–13] makes it possible to study the dependence of θ on external parameters proceeding from a microscopic picture, because it allows the properties of an inhomogeneous liquid to be calculated on the basis of particle-particle interactions.

In this work, the atomic fluid under a resonance electromagnetic irradiation is considered. The radiation frequency corresponds to the energy of atomic excitation, so that some atoms can be in the excited electron state. The lifetime of atoms in the excited electron state is much longer than the equilibrium establishment time in the system by means of the translational degrees of freedom. As a result, the fluid reveals equilibrium properties at a given concentration of excited atoms [14–22]. On the other hand, there appear new effective long-range (resonance dipole-dipole) interactions in a system composed of identical atoms in different electron states. They can change the macroscopic equilibrium properties of the system, even if the concentration of excited atoms is low [14–31]. In this connection, a number of experimental studies concerning fluids under a resonance irradiation conditions can be mentioned [32–42].

The microscopic theory of a fluid consisting of two-level atoms was proposed in our works [23–31]. This theory makes it possible to analyze the influence of an external electric field or resonance irradiation on such macroscopic equilibrium properties of a non-uniform system as the binodal, spinodal, coefficient of surface tension, and nucleation barrier for the vapor–liquid or liquid–vapor phase transitions. In our works, we used the density functional method both in the mean-field approximation and going beyond it.

In our recent work [43], we studied the effect of an external electric field on the wetting angle for a two-level atomic liquid. In this work in a similar way, we considered the resonance irradiation effect [14–22]. The structure of the work is as follows. First of all, the choice of a functional for the grand thermodynamic potential, which depends on the concentration of excited atoms, is discussed. The expression obtained for the grand thermodynamic potential makes it possible to find the surface tension, and, afterward with the use of the Young equation, the equilibrium wetting angle θ. The results obtained demonstrate that the growth in the concentration of excited atoms increases the wetting temperature T_w, i.e. the temperature, at which θ vanishes ($\cos\theta = 1$), and increases the wetting angle θ at a fixed temperature. Moreover, it can transform the complete wetting into a partial one and even stimulate the transition from the hydrophilic to hydrophobic state, when $\cos\theta$ becomes negative.

2. Density Functional Method

When developing the theory of an inhomogeneous fluid consisting of two-level atoms [23–31, 43], we use the density functional method [5–7]. The short-range repulsion was taken into account in the local approximation, which corresponds to the Carnahan–Starling equation of state for hard balls, whereas the long-range interaction was considered in the nonlocal approximation. Let us start from the following functional for the grand thermodynamic potential [23–31]:

$$\Omega[\rho(\mathbf{r})] = F_{\mathrm{CS}}[\rho(\mathbf{r})] + \\ + \frac{1}{2} \int\limits_{|\mathbf{r}_1-\mathbf{r}_2|\ge\sigma} d\mathbf{r}_1 d\mathbf{r}_2\, \rho^{(2)}(\mathbf{r}_1,\mathbf{r}_2)\, U(\mathbf{r}_1,\mathbf{r}_2) + \\ + \int d\mathbf{r}_1 \rho(\mathbf{r}_1)(V(\mathbf{r}_1) - \mu). \qquad (1)$$

The first term in this expression makes allowance for the short-range repulsion between hard balls with diameter σ written in the Carnahan–Starling form:

$$F_{\mathrm{CS}}[\rho(\mathbf{r})] = kT \int d\mathbf{r}_1\, \rho(\mathbf{r}_1) \Bigg(\ln(\Lambda^3\rho(\mathbf{r}_1)) + \\ + \frac{-1 + 6v\rho(\mathbf{r}_1) - 4v^2\rho^2(\mathbf{r}_1)}{(1 - v\rho(\mathbf{r}_1))^2} \Bigg), \qquad (2)$$

where μ is the chemical potential, Λ the thermal de Broglie wavelength, and $v = \pi\sigma^3/6$. The second term in Eq. (1) takes the long-range interaction into account. The pair distribution function $\rho^{(2)}(\mathbf{r}_1,\mathbf{r}_2)$ looks like

$$\rho^{(2)}(\mathbf{r}_1,\mathbf{r}_2) \simeq \rho(\mathbf{r}_1)\rho(\mathbf{r}_2) \qquad (3)$$

in the mean-field approximation. The attractive part of the long-range potential of pair interaction is given by the formula (see works [20–22])

$$U(\mathbf{r}_1,\mathbf{r}_2) = U(|\mathbf{r}_1 - \mathbf{r}_2|) = -\frac{3\sigma^3 a}{2\pi}\, \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|^6}. \qquad (4)$$

If there are excited atoms, the coefficient a depends on the concentration of atoms in the excited state, c_1, and the temperature T as follows:

$$a = \frac{v(E_1 - E_0)\alpha^2}{32} \left(1 - 2c_1 + 2(1 - c_1)c_1 \frac{E_1 - E_0}{kT}\right), \qquad (5)$$

where $E_1 - E_0$ is the energy of two-level atom excitation, $\alpha = |\mathbf{p}|^2/(\sigma^3(E_1 - E_0))$ is a dimensionless parameter characterizing the atom, and $|\mathbf{p}|$ the magnitude of the electric dipole moment for the transition between the ground and excited states of the atom (see works [20–22]).

The equation for the equilibrium density $\rho(\mathbf{r})$ is obtained from the condition $\delta\Omega[\rho(\mathbf{r})]/\delta\rho(\mathbf{r}) = 0$ [5–13] and looks like [23–31, 43]

$$kT \left(\ln\left(\Lambda^3\rho(\mathbf{r})\right) + \frac{8v\rho(\mathbf{r}) - 9v^2\rho^2(\mathbf{r}) + 3v^3\rho^3(\mathbf{r})}{(1 - v\rho(\mathbf{r}))^3} \right) + \\ + \int\limits_{|\mathbf{r}_1-\mathbf{r}|\ge\sigma} d\mathbf{r}_1 \rho(\mathbf{r}_1) U(|\mathbf{r}_1 - \mathbf{r}|) + V(\mathbf{r}) - \mu = 0. \qquad (6)$$

For a homogeneous fluid ($\rho(\mathbf{r}) = \rho$ and $V(\mathbf{r}) = 0$), the equation of state in terms of dimensionless variables reads

$$\frac{\pi}{\tau} = \eta \frac{1 + \eta + \eta^2 - \eta^3}{(1 - \eta)^3} - \frac{a(c_1,\tau)}{\tau}\eta^2.$$

Here,

$$a(c_1,\tau) = \frac{\alpha^2}{32}\left(1 - 2c_1 + \frac{2(1 - c_1)c_1}{\tau}\right), \qquad (7)$$

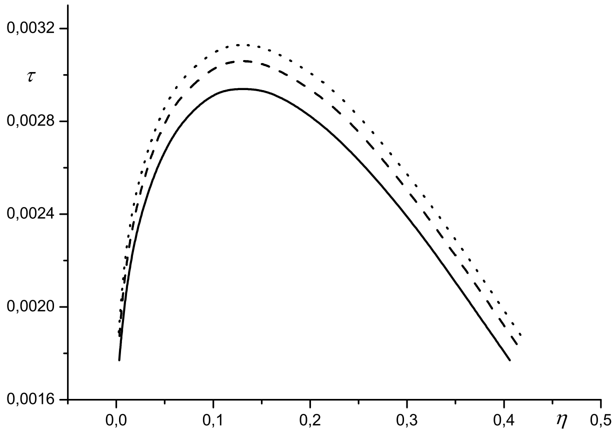

Fig. 1. Binodals in the density–temperature plane for various concentrations of excited atoms $c_1 = 0$ (solid curve), 0.00006 (dashed curve), and 0.0001 (dotted curve)

$\pi = pv/(E_1 - E_0)$ is the dimensionless pressure, $\tau = kT/(E_1 - E_0)$ is the dimensionless temperature, and $\eta = v\rho$ is the dimensionless density. Hereafter, for simplicity, we put $\alpha = 1$ [1].

From Eq. (7), it is easy to determine the critical parameters – namely, the critical density, temperature, and pressure – and to plot the corresponding phase diagram (see Fig. 1). Since the parameter a in Eq. (7) depends on the concentration of excited atoms c_1, the critical temperature τ_c and the critical pressure π_c also depend on c_1.

Now, we have to know the explicit expression for the external potential $V(\mathbf{r})$ that describes the interaction between the solid surface (substrate) and the atoms in the liquid. For simplicity, we assume that a continuous solid surface, $z < 0$, is formed by uniformly distributed two-level atoms with the density ρ_s. Those atoms interact with the environment of two-level atoms by means of the same potential as in the fluid (see, e.g., works [46–48]) [2]. The contribution of a semiinfinite flat solid surface to $V(x, y, z) = V(z)$ at $z \geq \sigma$ is calculated by integrating the interatomic interaction $-3\sigma^3 a/(2\pi R^6)$ [see Eq. (8)]. As a result, we obtain

$$V(z) = \rho_s \times$$

$$\times \int\limits_{-\infty}^{\infty} \mathrm{d}x' \int\limits_{-\infty}^{\infty} \mathrm{d}y' \int\limits_{-\infty}^{0} \mathrm{d}z' U(\sqrt{x'^2 + y'^2 + (z - z')^2} =$$

$$= -\frac{\rho_s\, a\, \sigma^3}{4}\frac{1}{z^3}. \tag{8}$$

Hereafter, for convenience, we put $\eta_s = \rho_s v = 1$. Additionally, we put $V(z) = \infty$ at $0 \leq z < \sigma$.

Substituting the solution of Eq. (6) for the equilibrium density into Eq. (1), we obtain the required expression for the grand thermodynamic potential of the inhomogeneous system, Ω.

3. Results and Their Discussion

The wetting angle θ is determined as follows. Firstly, the liquid-vapor surface tension γ_{lv} is calculated. We put $V(\mathbf{r}) = 0$ in Eq. (6) and find a solution for the equilibrium density $\rho(z)$ depending on the height z. Substituting this solution into formula (1), we obtain an expression for the grand thermodynamic potential of a two-phase fluid, Ω. The surface tension γ_{lv} is determined from the relation $\Omega = \Omega_0 + \gamma_{lv} S$, where $\Omega_0 = -pV$ is the grand thermodynamic potential for the homogeneous system in the volume V (the pressure in the system is p in this case), and S is the interphase surface area.

The surface tension at the solid-liquid, γ_{sl}, or solid-vapor, γ_{sv}, interface is calculated in the framework of the same scheme, but the interaction potential $V(\mathbf{r})$ (8) has to be taken into consideration at that. We assume that the interface is located at $z = 0$. While calculating γ_{sl}(or γ_{sv}), we put the liquid (vapor) density at z to be equal to the corresponding value at $z \to \infty$, i.e. $\rho(z) = \rho_l$ or $\rho(z) = \rho_v$, respectively. Afterward, we determine the equilibrium density $\rho(z)$ for all $0 \leq z < \infty$, the grand thermodynamic potential Ω, and, finally, the value for γ_{sl} or γ_{sv}. The wetting angle θ is determined from the Young equation

$$\gamma_{sv} - \gamma_{sl} - \gamma_{lv} \cos\theta = 0. \tag{9}$$

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[1] Of course, for real atoms, the value of α differs from unity. For instance, for the $1s^2 - 1s2p$ transition in a He atom, $E_1 - E_0 \approx 21.27$ eV, $|\mathbf{p}|^2/\sigma^3 \approx 0.15$ eV, so that $\alpha \approx 0.007$. At the same time, for the $6s^2 - 6s6p$ transition in a Ba atom, $E_1 - E_0 \approx 2.24$ eV, $|\mathbf{p}|^2/\sigma^3 \approx 0.94$ eV, and $\alpha \approx 0.420$. For those calculations, we used data from [44] (see also work [45]). The analysis of parameter for other systems, which would be of interest for experimental studies, is outside the scope of our work.

[2] If the solid wall is formed by uniformly distributed two-level atoms located in the plane $z = 0$, we obtain the dependence $V(r) \sim 1/z^4$ rather than $V(r) \sim 1/z^3$ as in Eq. (8).

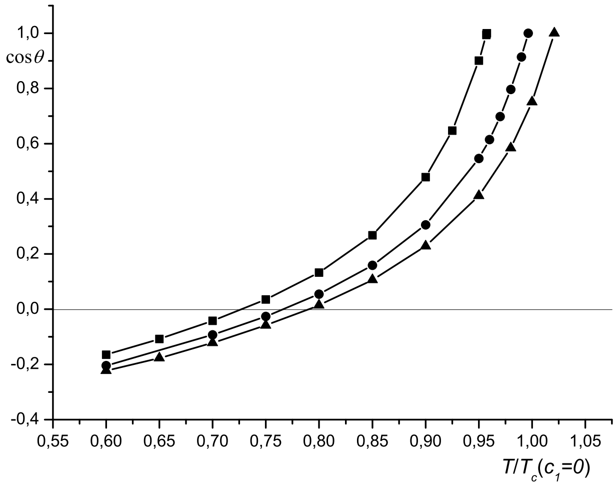

Fig. 2. Dependences of $\cos\theta$ on the temperature $T/T_c(c_1 = 0)$ for various concentrations of excited atoms $c_1 = 0$ (squares), 0.00006 (circles), and 0.0001 (triangles)

This is a final stage of finding $\theta(T, c_1)$ from the first principles, i.e. proceeding from the interparticle interactions modified by the resonance irradiation. The results of calculations are depicted in Figs. 2 and 3. The temperature dependence of the wetting angle exhibited in Fig. 4 satisfies the relation

$$1 - \cos\theta \propto (T_w - T)^{2-\alpha_s}$$

with $\alpha_s = 1$, which corresponds to the wetting phase transition of the first order [49].

Let us discuss the results obtained. In the case of a fluid of two-level atoms, the presence of excited atoms increases the long-range attraction. As a result, the critical temperature of the liquid increases: $T_c(c_1 \neq 0) > T_c(c_1 = 0)$. Hence, after the irradiation has been "switched-on" at a constant temperature, the two-phase state "turns out" farther from the critical region. The surface tensions γ_{sl} and γ_{sv} increase with the concentrations of excited atoms. If γ_{sl} grows more rapidly than γ_{sv}, $\cos\theta$ may can change its sign in accordance with formula (9), and the hydrophilic surface ($\cos\theta > 0$) becomes hydrophobic ($\cos\theta < 0$).

We adopted a very simple model for the substrate. The external potential $V(\mathbf{r})$ can be smaller, e.g., owing to the reduction of η_s. In this case, the substrate role decreases: it remains hydrophobic even in the absence of excited atoms. The wetting angle increases with the concentration of excited atoms c_1. If the temperature is very close to but remains a bit lower than the complete wetting temperature $T_w < T_c$, even a low concentration of excited atoms c_1 can induce noticeable changes of θ (see the curve for $\tau = 0.00282$ in Fig. 3).

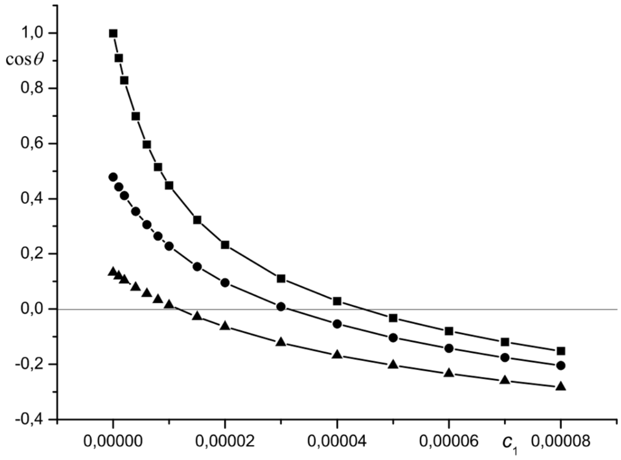

Fig. 3. Dependences of $\cos\theta$ on the concentration of excited atoms c_1 at various temperatures $\tau = 0.9574\tau_c(c_1 = 0) = 0.00282$ (squares), $\tau = 0.9\tau_c(c_1 = 0) = 0.00265$ (circles), and $\tau = 0.8\tau_c(c_1 = 0) = 0.00236$ (triangles)

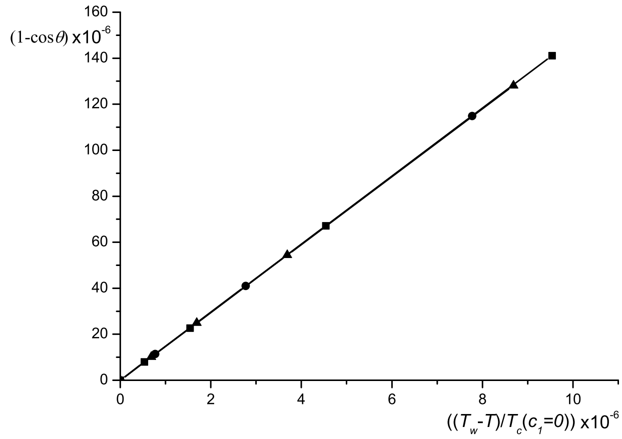

Fig. 4. Dependence of $1 - \cos\theta$ on the temperature $(T_w - T)/T_c(c_1 = 0)$. The dependence $1 - \cos\theta \propto (T_w - T)^{2-\alpha_s}$ with $\alpha_s = 1$ describes the phase transition of the first kind. The dependences for three different concentrations of excited atoms c_1 lie on the same straight line: $c_1 = 0$ (squares), 0.00006 (circles), and 0.0001 (triangles)

To summarize, we applied the density functional method to a simple fluid of two-level atoms in order to study the influence of the external irradiation on the wetting properties. The corresponding theoretical analysis could describe liquid droplets that are on a solid surface in equilibrium with vapor, when the system is subjected to the resonance irradiation. Excited atoms, as was in the case of external electric field [43], enhance the interatomic attraction at large distances. However, since the parameter a [Eq. (5)] or $a(c_1, \tau)$ [Eq. (7)] depends on the temperature, there is no universality inherent to the case with the electric field (recall that the wetting angle θ expressed in terms of $T/T_c(\mathcal{E})$, where $\mathcal{E}$ is the dimensionless electric field [43], does not depend on the electric field). The appearance of excited atoms at temperatures slightly below the temperature $T_w(c_1 = 0) < T_c(c_1 = 0)$ can significantly increase the wetting angle θ and even stimulate the transition from hydrophilicity to hydrophobicity.

We hope that our theoretical analysis will stimulate the interest of experimenters to the study of the influence of a resonance irradiation on the solid surface wetting by liquid. Information concerning similar researches of the influence of an external electric field on the wetting can be found in works [50–55].

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Received 02.07.17.
Authorized translation from Ukrainian by O.I. Voitenko

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ЗМОЧУВАННЯ В УМОВАХ РЕЗОНАНСНОГО ЕЛЕКТРОМАГНІТНОГО ОПРОМІНЕННЯ

Резюме

Обговорюється вплив резонансного електромагнітного опромінення на змочування рідиною твердої поверхні. Розглядається проста модель плину дворівневих атомів, для яких зміни міжатомних взаємодій за наявності зовнішнього резонансного опромінення можуть бути знайдені за допомогою квантово-механічної теорії збурень. Побудувавши функціонал великого термодинамічного потенціалу, з допомогою методу функціонала густини розраховується поверхневий натяг рідина–пара, тверда поверхня–рідина, тверда поверхня–пара і з рівняння Юнга знаходиться кут змочування. Показано, що наявність резонансного опромінення може помітно збільшувати кут змочування.