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Physics Letters A 361 (2007) 252–256
PHYSICS LETTERS A
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Nano liquid-metal fluid as ultimate coolant

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Received 10 July 2006; received in revised form 9 September 2006; accepted 18 September 2006

Communicated by R. Wu

Abstract

We proposed for the first time the concept of the nano liquid-metal fluid, aiming to establish an engineering route to make the highest conductive coolant. Using several widely accepted theoretical models for characterizing the nano fluid, the thermal conductivity enhancement of the liquid-metal fluid due to addition of more conductive nano particles was predicted. Further, the effects of particle size, cluster of nano particle, solid-like layer due to adsorption, volume fraction and particle types were evaluated. Having the highest conductivity, being electromagnetically drivable, the liquid metal with low melting point is expected to be an idealistic base fluid for making super conductive solution which may lead to the ultimate coolant in a wide variety of heat transfer enhancement area.

Keywords: Chip cooling; Thermal management; Liquid metal; Nano fluid; Ultimate coolant; Nano liquid metal flow; Heat transfer enhancement; Base fluid

The last two decades witness an explosive growth of personal computers and workstations in the microelectronic industry. Meanwhile, the chip integration density is approaching its limit due to "thermal barrier" encountered. As is well-known, over-heating of a computer chip may result in shortened life, malfunction, low reliability and failure of work. Therefore, removal of the large amount of heat generated in the electronic components remains as a big challenge facing current computer system designers and thermal management engineers. Compared with the widely adopted air-cooling, liquid-cooling appears much efficient at drawing heat away from the processor and is therefore becoming a major stream in developing new generation chip cooling device. For example, with the forced convective flow of water, an improvement in heat transfer capacity over air-cooling has been found to be more than a factor of 10 [1]. However, such method also has its limitation. The small thermal conductivity of water would lower its effectiveness as a cooling fluid. Therefore, researchers are considering enhance the convective heat transfer effect of water by adding highly conductive nano particles such as copper or aluminum into the solution suspension [2,3], which leads to the recent explosive investigation on the nano fluids. Compared with the suspended particles of millimeter-or-micrometer dimensions, nano fluids show better stability and rheological properties, dramatically higher thermal conductivities, and no penalty in pressure drop [4]. Even a small amount ($<1\%$ volume fraction) of Cu nano particles or carbon nano tubes dispersed in ethylene glycol or oil is found to increase the inherently poor thermal conductivity of the liquid by 40% and 150%, respectively [2,5,6]. Traditional nano fluid is generally made by dispersing nano particles in an ordinary liquid such as water, ethylene glycol or oil. However, such improvement is still rather limited due to the used base fluids. Particularly, the mixed solution may easily subject to additional troubles during thermal management such as susceptibility to fouling, particle deposition or conglomeration, degeneration of solution quality and flow jamming over the channels, etc.

Compared with the ordinary liquid such as water or other organic fluids, metal has a much higher thermal conductivity, no matter what state it stays at. Thus, if using certain liquid metal or its alloys with low melting point as the coolant, a much wider cooling capacity will be reached. Starting from this point, Liu and Zhou [7,8] introduced to adopt liquid metal or its alloy to cool the computer chip in the early year of 2002. The recent

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doi:10.1016/j.physleta.2006.09.041

work by Miner and Ghoshal [9] further strongly support this effort. Clearly, with about several dozen times larger thermal conductivity than that of water, the liquid metal such as gallium or mercury shows a very good property in transferring heat away. It is among the naturally existing liquid to have the highest conductivity however the lowest melting point. Until now, one would still ask such an intriguing question, i.e. is there any ultimate coolant which has the highest conductivity or could we make things better in improving the liquid metal? It is from this serious consideration, we proposed in this study to further significantly magnify the conductivity of the liquid metal coolant by adding nano particles with superior conductivity to the liquid metal or its alloy. This promising route may lead to making the highest thermally conductive fluid. Several typical pictures actually taken for such liquid or its components were depicted in Fig. 1. Here, we will disclose the basic features of the conductivity enhancement of the nano liquid-metal fluid, based on several well accepted theoretical models for characterizing the nano fluids.

As one knows, the gallium and mercury are two typical materials which in most cases would stay in liquid state near the room temperature. Especially, the liquid gallium satisfies such merits: non-poisonous, non-caustic for most materials, low viscosity, and high thermal conductivity. If the surrounding temperature becomes low enough to overcome the super cooling point of the liquid gallium, it would become solidified and is not valid as a coolant. Compared with this, the mercury has a much lower melting point, say −38 °C. Therefore, in most of the commonly encountered situations, it would always keep staying in a liquid state. The only trouble of the mercury comes from its toxicity. As a typical naturally existing metal with extremely low melting point, these two materials will be theoretically evaluated when used as the base fluid for making nano liquid-metal fluid. Their comparison in particular thermal features with the traditional fluids such as water and ethylene glycol can be found in Table 1. With an extremely high conductivity however relatively small viscosity, the liquid metal could serve as an idealistic base solution for making highly conductive nano fluid. This can be explained a little more by the following analysis.

Here, the nano liquid-metal was proposed as a highly conductive heat transfer fluid, which is superior over many existing base liquid such as water. The evaluation as performed in [9] strongly supports such plausible effort. Our group had also experimentally compared the temperature responses at the simulated hot chip and the radiator, when using liquid metal or water as coolant, respectively [8,10]. All the results indicate that, the liquid metal has a better heat transfer performance than that of water. Under the same situation, the liquid metal cooling results in a higher radiator temperature, which significantly improves the heat transfer between the radiator and the surrounding air. Therefore, a lower temperature for the simulated hot chip will

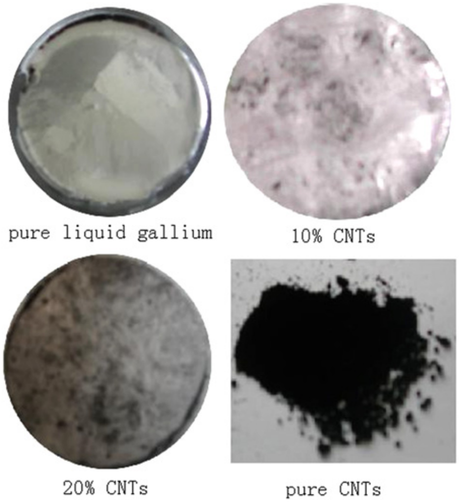

Fig. 1. Carbon nano tubes are added in the liquid gallium.

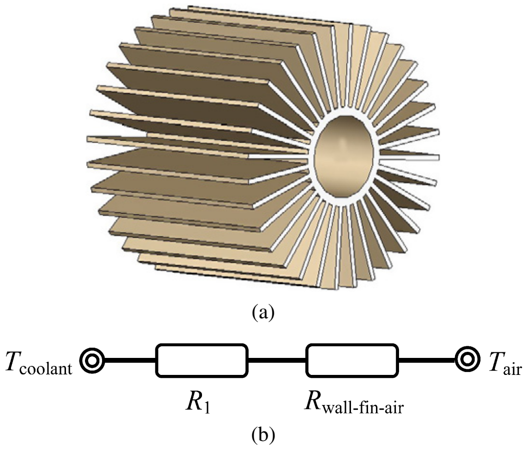

Fig. 2. Heat transfer for flowing liquid running inside a tube radiator (T_{coolant} and T_{air} are temperatures for coolant and surrounding air, respectively). (a) Cylindrical tube radiator with fin structure. (b) Thermal resistance between coolant and ambient air.

Table 1
Physical properties for liquid metals or traditional fluids

Liquid metals or traditional fluids	Thermal conductivity W/(m K)	Heat capacity J/(kg K)	Density kg/m^3	Surface tension mN/m	Viscosity $\times 10^8$ m^2/s	Melting point °C	Boiling point °C
Gallium	28.7[a]	381.5[c]	6095 [c]	709 [c]	29.7[d]	29.8	2200
Mercury	8.4[b]	138.2[e]	13470 [e]	483 [f]	10.4[e]	−38	357
Water	0.6	4183	1000	72.8	100.6	0	100
Ethylene glycol	0.258	2349	1132	48.4	18.86	−12.6	197.2

Data measured:
[a] at 77°C; [b] at 50 °C; [c] at 30 °C; [d] at 32 °C; [e] at 50 °C; [a] at 20 °C.

be obtained, which is just the core target for computer thermal management.

Such finding also has its theoretical root which can be illustrated through a simplified analysis as given below. Without loosing any generality, if taking the heat transfer of liquid metal running inside a cylindrical tube radiator (see Fig. 2(a)) with fins for example, the whole thermal resistance from the coolant, i.e. the liquid metal or the water, to the ambient air can be represented in Fig. 2(b).

The total thermal resistance between coolant and air can be expressed as:

$$R = R_1 + R_{\text{wall-fin-air}} \tag{1}$$

where, R_1 is the thermal resistance between the coolant and the inner wall of the tube radiator and $R_1 \sim 1/h$ (h is convective heat transfer coefficient between coolant and tube wall). $R_{\text{wall-fin-air}}$ represents the total thermal resistance between the wall, fin of the tube and the surrounding air. For the same radiator working in the same ambient environment, $R_{\text{wall-fin-air}}$ would be the same no matter what coolant is used. Therefore, the total thermal resistance between the coolant and air depends mainly on R_1. From many previous studies on liquid metal heat transfer such as those performed on nuclear reactor, it has been well known that the convective heat transfer coefficient h between the liquid metal coolant and its surrounding wall is much larger than that between the water and the wall. This will therefore result in a smaller thermal resistance R_1. And, a stronger heat transfer performance over water can then be obtained using liquid metal as the coolant.

A simplified equation can be used to characterize the heat transferred via coolant as

$$Q = \frac{T_{\text{coolant}} - T_{\text{air}}}{R}. \tag{2}$$

Compared with the water cooling case, the temperature at the radiator is higher with the flow of the much hotter liquid-metal coolant. Besides, the overall thermal resistance for liquid-metal cooling is smaller. Therefore, one can arrive at the conclusion that more heat Q will be dissipated when the liquid metal is adopted as heat transfer coolant in chip cooling. Clearly, the higher value for T_{coolant} and the smaller overall thermal resistance R, the more heat it will dissipate to the environment. In this sense, if a more conductive fluid such as nano liquid metal was further used to replace the liquid metal, a much significant heat dissipation capacity for the cooling chip can still be further realized. This is a core reason for us to introduce the nano liquid metal as a much better base fluid than water. Since such coolant has the highest thermal conductivity among various liquids existing in nature, its adoption into the chip cooling area or even nuclear reactor would have significant value for developing a highly efficient heat transport device.

As reflected by Table 1, the traditional fluids have two disadvantages in developing a nano fluid. One is their rather small thermal conductivity. Another lies in the lager density difference between the nano particles and the base fluid. For the liquid metal used as coolant, its large surface tension would tend to prevent the nano particles from easily depositing. The much smaller surface tension of traditional fluid generally limits the maximum volume fraction for the addition of nano particles. If nano particles were over loaded, they would easily deposit to the bottom of the base fluid. Therefore, the nano fluids used for the purpose of enhanced heat transfer are often dilute multi-component fluids and the volume fractions of nano particles are generally below 5–10%. Therefore, only a very limited improvement on the thermal conductivity can be obtained. Compared with this, the liquid-metal appears much conductive as an idealistic base fluid. Besides, its surface tensions can have approximately 7–10 times larger than that of water and ethylene glycol. As a result, a much larger volume fraction of nano particles can be added to the liquid metal (see Fig. 1). In this way, more capacity for enhancing the effective thermal conductivity of the nano fluid will be expected.

Previous efforts demonstrate that several typical models as listed in Table 2 are approximately true for characterizing the effective thermal conductivity of nano fluids. Although these models may deviate from the experimental data more or less, they still provide a very useful way in theoretically predicting the tendency of enhancement of thermal conductivity in nano fluids. Among these, for low particle-concentration suspension, the Bruggeman model shows almost the same result as the Maxwell–Garnett self-consistent approximation (MG model) [11] will give. For a particle percolation situation or when the particle concentration is sufficiently high, the MG model fails to predict precisely the experimental results, while the Bruggeman model can still fit well with experimental data. Up to now, there are no existing correlations for effective thermal conductivity of liquid metal. As a first and intuitive step towards probing into the thermal properties of the nano liquid-metal fluid, some of these widely accepted models will be used to predict the effective thermal conductivity enhancement of the nano liquid-metal fluid. And a few typical results obtained are displayed in Figs. 3 and 4.

In the calculation process, the thermal conductivities of carbon-nanotube, gold, silver, copper are fitted as 3000 W/(m K), 315 W/(m K), 427 W/(m K), and 386 W/(m K), respectively. We do not consider aluminium nano particle because experiments have shown that it would be corroded by the liquid gallium with the time passing on. Further, because liquid metal has a much larger surface tension than that of traditional fluid, a maximum volume fraction up to 20% was considered for the addition of nano particles. In fact, experimental tests show that even a volume far larger than this can still easily guarantee the liquid state of the nano fluids. The actual photos taken for different volume fraction of Carbon Nano Tubes (CNTs) in liquid gallium are depicted in Fig. 1. Although the mixing is not as easy as that in water, it can be found that the mixture keeps staying in liquid state even when the added volume faction is up to 20%.

Further, the nano particles were all treated as spheres. Therefore the Hamilton–Crossor model gives the same results as that of the Maxwell model. Bruggeman model may predict a little more real result due to taking into consideration the clusters of nano particles. Clearly, a larger tendency for the enhancement of nano liquid-metal fluid has been indicated in Fig. 3

Table 2
Typical models for effective thermal conductivity of nano fluids. k_{eff}, effective thermal conductivity of solid/liquid suspensions; k_f, thermal conductivity of base fluid; k_p, thermal conductivity of nano particle; $\alpha = k_p/k_f$, thermal conductivity ratio; $\beta = (\alpha - 1)/(\alpha + 2)$; n, particle shape factor; and v, particle volume fraction

Models	Expressions	Remarks
Maxwell [11]	$\frac{k_{\text{eff}}}{k_f} = 1 + \frac{3(\alpha-1)v}{(\alpha+2)-(\alpha+1)v}$	Spherical particles are considered.
Hamilton–Crossor [12]	$\frac{k_{\text{eff}}}{k_f} = \frac{\alpha+(n-1)-(n-1)(1-\alpha)v}{\alpha+(n-1)+(1-\alpha)v}$	Spherical and non-spherical particles are considered: $\alpha = 3$ for spheres, $\alpha = 6$ for cylinders.
Jeffrey [13]	$\frac{k_{\text{eff}}}{k_f} = 1 + 3\beta v + \left(\frac{15}{4}\beta^2 + \frac{9}{16}\beta^3 \frac{\alpha+2}{2\alpha+3} + \cdots\right)v^2$	High-order terms represent pair interaction of randomly dispersed spheres.
Davis [14]	$\frac{k_{\text{eff}}}{k_f} = 1 + \frac{3(\alpha-1)}{(\alpha+2)-(\alpha+1)v}[v + f(\alpha)v^2 + o(v^3)]$	High-order terms represent pair interaction of randomly dispersed spheres. $f(\alpha) = 2.5$ for $\alpha = 10$, $f(\alpha) = 0.5$ for $\alpha = \infty$.
Lu–Lin [15]	$\frac{k_{\text{eff}}}{k_f} = 1 + av + bv^2$	Spherical and non-spherical particles are considered.
Bruggeman [16]	$\frac{k_{\text{eff}}}{k_f} = \frac{1}{4}\{(3v - 1)k_1 + [2 - 3v] + \sqrt{\Delta}\}$ $\Delta = (3v-1)k_1^2 + [2-3v]^2 + 2[2+9v(1-v)]k_1$	The clustering of nano particles and the surface adsorption are considered.
Bonnecaze–Brady [17]	Numerical simulation	Near- and far-field interactions among two or more particles are considered.

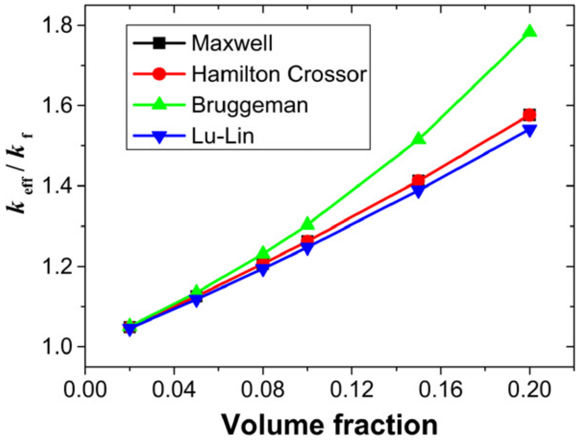

Fig. 3. Thermal conductivity enhancement ratio as a function of volume fraction for nano copper in liquid-gallium suspensions using different model.

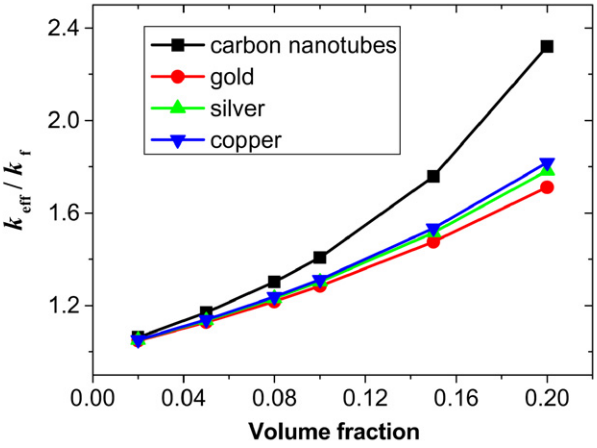

Fig. 4. Thermal conductivity enhancement ratio as a function of volume fraction for different nano particles in liquid-gallium suspensions.

for all the models. The results most probably true will fall in the area surrounded by the two boundary curves. From Fig. 4, one can still find the effect of thermal conductivity of several typical nano particles on the effective thermal conductivity of nano liquid-metal fluid using Bruggeman model. The thermal conductivity enhancement ratio can even reach 2.3 if the carbon nano tube is dispersed in the liquid gallium with a volume fraction 20%. As a comparison, addition of other nano particles including gold, silver and copper into the liquid gallium also realizes a thermal conductivity enhancement ratio of more than 1.6. Considering both the high thermal conductivity of the base fluid and the large volume fraction, this nano fluid made from the liquid-metal shows a very promising future for the thermal management application in super-CPU chip cooling or the situations request seriously high heat flux removal. The similar results as that in both Figs. 3 and 4 can also be obtained for the case of using mercury as the base liquid metal fluid.

It should be pointed out that, the above models do not take into concern the size effect on the effective thermal conductivity. In fact, this does play a vital role. For nano fluids, researchers have found that the size and shape, the clustering of nano particles and the surface adsorption could be the major reason of enhancement [16], while the Brownian motion of nano particles contributes much less than other factors [18]. It has been noted before that concave geometry of the liquid bridge occurs when the liquid is wetting the particles. This would result in an attractive force leading to the attachment of some molecules of base fluid to the nano particles. Recent experimental study has shown that molecules of normal liquids close to a solid surface organize into layered structures much like a solid [19]. Theoretical investigations have also suggested that a solid-like ordering of suspended spheres will occur in the confined multi-phase contact region at the edge of the spreading fluid over the nano particle [20,21]. Furthermore, there is evidence that such solid-like structure of a liquid at the surface is a governing factor in heat conduction from a solid wall to an adjacent liquid [22]. For the same material, the conductivity in solid state is higher than that in liquid state. The thermal conductivity can thus be enhanced when the surrounding molecules of base fluid are attached to the nano particle. Taking the adsorption of

Table 3
Thickness of liquid layer (unit: nm)

Base fluid	Water	Ethylene glycol	Gallium	Mercury
Thickness	2.84	4.15	2.44	2.66

liquid molecules on the particle surface as a monolayer shell, its thickness can be expresses as [23]

$$h = \frac{1}{\sqrt{3}}\left(\frac{4M}{\rho_f N_A}\right)^{\frac{1}{3}} \tag{3}$$

where, M is the molecular weight of base liquid, ρ_f the density of liquid, and N_A is Avogadro constant (6.02×10^{23}/mol).

From Eq. (3), we know that the thickness is only related to the base fluid. However, more complex situation should exist which needs further clarifications in the near future. Table 3 lists the thickness of different base fluid. Yu and Choi [24] calculated thermal conductivity enhancement ratio as a function of particle radius for copper-in-ethylene-glycol suspensions when volume fraction of nano particle is 1%.

Based on effective medium theory, the apparent thermal conductivity k_{app} of the equivalent particles can be modified as

$$k_{\text{app}} = \frac{2+\alpha+2(\alpha-1)\lambda}{2+\alpha-(\alpha-1)\lambda} k_{\text{shell}} \tag{4}$$

where, $\alpha = k_p/k_f$, $\lambda = [d/(d+2h)]^3$, and k_{shell} is the thermal conductivity of solid-like shell surrounding the nano particles, ranging from thermal conductivity of base fluid to that of corresponding solid state; d is the diameter of nano particle.

The equivalent volume fraction can easily be obtained by

$$v_{\text{app}} = \frac{v}{\lambda}. \tag{5}$$

Using Eqs. (3), (4) and (5), one can get the modified Maxwell model as follows

$$\frac{k_{\text{eff}}}{k_f} = 1 + \frac{3(\alpha_{\text{app}}-1)v_{\text{app}}}{(\alpha_{\text{app}}+2)-(\alpha_{\text{app}}+1)v_{\text{app}}}. \tag{6}$$

As indicated in Fig. 5 by calculating Eq. (6), the thermal conductivity enhancement ratio decreases dramatically when the sizes of nano particle increase. When the diameter of nano particle is larger than 80 nm, only a slight enhancement can be obtained. That is because the ratio $2h/d \ll 1$, the volume fraction of equivalent nano particles can increase slightly. For simplicity, Fig. 5 shows the effect of size only considering that the nano particles are balls. In fact, the shape of nano particles also plays a vital role in the thermal conductivity enhancement in nano fluids. For example, carbon nano tubes in fluid can enhance the thermal conductivity more than other nano particles in nano fluid [14]. Further evaluation on such problem can also possibly be made using some existing theoretical models and their modified forms. Clearly, the concept of nano liquid-metal fluid opens a promising way for making a highly conductive coolant.

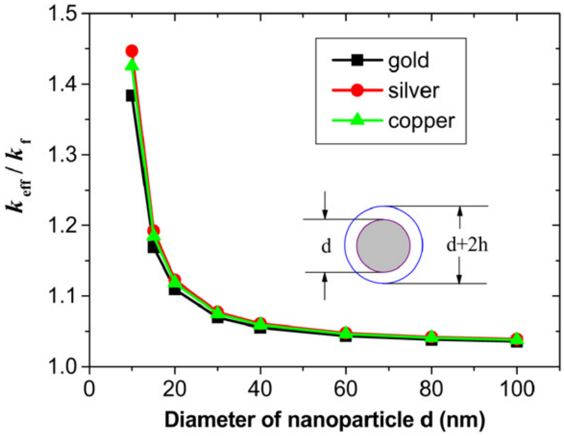

Fig. 5. Thermal conductivity enhancement ratio as a function of diameter of different nano particles in liquid-metal suspensions when volume fraction of nano particle in nano fluid is 10%.

Acknowledgements

This work is financially supported by the National Natural Science Foundation of China under Grants of 50575219, 50576103 and 50325622.

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