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A THEORETICAL CONSIDERATION OF DIRECTIONAL EFFECTS IN HEAT FLOW AT THE INTERFACE OF DISSIMILAR METALS

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Abstract—Several investigators have found that the resistance to heat transfer at certain metal–metal interfaces is dependent upon the direction of heat flow across these interfaces. This paper shows that such a phenomenon can be explained by application of the theory of heat conduction in the solid state.

NOMENCLATURE

A, contact area, cm^2;
C, numerical constant, $(4\pi mk^2/h^3) = 0{\cdot}75 \times 10^{21}$, (cm^2 sec °K^2)$^{-1}$;
E, energy of an electron, eV;
$\bar{E}$, average energy, eV;
E_0, height of potential barrier, eV;
E_w, work function, eV;
E_m, Fermi level, eV;
h, Planck's constant, $6{\cdot}624 \times 10^{-34}$ J-s;
$\bar{h}$, interfacial heat-transfer coefficient, chu/ft^2 h degK;
k, Boltzmann constant, $1{\cdot}381 \times 10^{-23}$ J/degK;
l, thickness of potential barrier, Å;
κ, $2m/k^2$, 0·251 (eV-Å^2)$^{-1}$;
m, mass of an electron, $9{\cdot}107 \times 10^{-31}$ kg;
N, number of electrons transferred from one metal to the other, m^{-2} s^{-1};
q, heat-flow rate, eV/m^{2}s;
T, temperature, °K;
$\bar{T}$, transmissivity, dimensionless;
v, velocity, linear units/s;
π, constant, 3·1416.

Subscripts

1, metal 1;
2, metal 2;
12, direction from metal 1 to metal 2;
21, direction from metal 2 to metal 1;
a, actual;
e, associated with electrons;
n, nominal;
p, associated with phonons;
s, oxide film;
t, total;
x, x-direction, direction of heat flow;
y, y-direction;
z, z-direction.

INTRODUCTION

IN 1936, Starr [1] conducted experiments with a copper–copper oxide rectifier which seemed to indicate that thermal conductivity at the interface between the two materials depended upon the direction of heat flow across the interface. These results were later described in a standard text on rectifiers [2]. However, in 1951, Horn [3] criticized Starr's experiments on the basis that Thomson e.m.f. caused by the temperature gradient across the rectifier led to spurious results, since Starr used un-insulated thermocouples with a common lead. In 1955, Barzelay *et al.* [4] found in the course of determining thermal conductivity of aircraft joints that the conductivity across the aluminum–stainless-steel joints depended on the direction of heat flow. Since their experiments were not specifically designed to test for the presence of this effect, they proposed further experimentation in the field.

Finally, Rogers and his group at the University of Bristol carefully designed experimental apparatus to determine whether the asymmetric heat-conduction effect really existed [5]. Rogers found a definite directional heat-transfer effect

in the systems he studied, but he offered no theoretical explanation for this phenomenon, although he did suggest that an interfacial potential barrier might be responsible.

THEORY

The directional heat-transfer phenomenon at the interface of dissimilar metals in a metal–metal contact can be predicted by application of the theory of heat conduction in the solid state.

When two metal surfaces are brought together, a direct metal-to-metal contact does not exist across the entire interface, and heat transfer across this interface may take place by several mechanisms. These are:

(1) electronic heat conduction;
(2) phonon heat conduction;
(3) radiative heat transfer (where metals are not in actual surface-to-surface contact);
(4) conduction across fluid film at interface;
(5) convection across fluid film at interface.

In his experiments, Rogers eliminated the last two modes of heat transfer by carefully cleaning surfaces and placing the entire system under a vacuum. Since heat transfer by radiation is negligible at the temperature of Rogers' experiments, only the first two mechanisms of heat transfer need be considered in a theoretical analysis of his data.

When many metals are exposed to air, they rapidly become coated with a thin film of oxide. For example, aluminum, one of the heat-transfer metals used in Rogers' work, is rapidly coated with an oxide film greater than 20 Å thick upon exposure to air. In heat transfer across an interface formed by a metal-oxide–metal contact, the oxide layer or layers can be considered a low-conductivity barrier layer at the surface of contact between two metals of different conductivity. After the physical contact which forms the interface, electrons may be thought of as "flowing" over the top of the insulating barrier until a double layer of charge is built up, bringing the Fermi energy levels of the metals to values such that the number of electrons "transferred" from metal 1 to metal 2 is equal to the number of electrons "transferred" in the opposite direction; i.e. we are at steady-state.

If metal 1 has a greater work function than metal 2, the electrons will flow from metal 2 to metal 1, since the electrons in the conduction band of metal 2 are nearer to the top of the potential barrier than those in metal 1. A diagram of energy levels is given in Fig. 1.

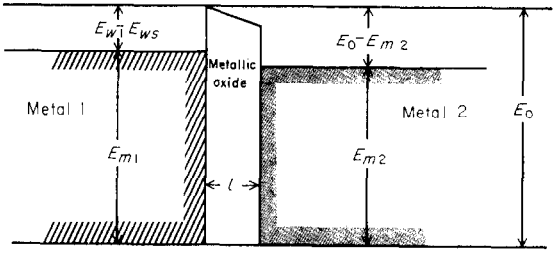

FIG. 1. Simplified diagram of steady-state electronic energy levels at the interface of two dissimilar metals with oxide layer; heat transfer taking place across the interface.

According to the principles of quantum mechanics, it is possible for an electron to penetrate a potential barrier, even though it possesses less energy than the height of the potential barrier. This effect is negligible in this case, as will be discussed later. In addition, not all the electrons in one metal which have a greater energy that the height of the potential barrier will transfer into the other metal. Thus, it is necessary to use quantum mechanics to calculate the transmission of electrons across such a potential barrier, and this will be done later in this paper.

The total heat transfer is expressed as follows:

$$q_t = q_e + q_p. \tag{1}$$

Considering only electronic heat transfer, and using a simple energy balance,

$$q_{e_{12}} = N_{12}\bar{E}_{12} - N_{21}\bar{E}_{21}. \tag{2}$$

At steady state, there is no net flow of electric current across the interface and therefore,

$$N_{12} = N_{21}. \tag{3}$$

Then, we can write

$$q_{e_{12}} = N_{12}(\bar{E}_{12} - \bar{E}_{21}). \tag{4}$$

N_{12} is calculated by use of the random-current-density model for electrons passing over a potential barrier, as developed by Richardson [6] for

quantitatively describing thermionic emission. For the model developed in this paper,

$$N_{12} = \frac{A_{a_{12}}}{A_n} \bar{T}_{12} \frac{2m^3}{h^3} \int_{v_x}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{1}{1 + \exp(E - E_{m_1}/kT_1)} v_x \, dv_x \, dv_y \, dv_z, \quad (5)$$

where $v_x = (2E_0/m)^{\frac{1}{2}}$.

Since $(E - E_{m_1})/kT_1 \gg 1$, we can neglect the term unity in the denominator of the integrand. Integrating, we obtain,

$$N_{12} = \frac{A_{a_{12}}}{A_n} \bar{T}_{12} T_1^2 \frac{4\pi mk^2}{h^3} \exp - \left(\frac{E_{w_1} - E_{w_2}}{kT_1}\right). \quad (6)$$

The average energy carried by an electron is given by the relationship

$$\bar{E} = \frac{\text{total energy carried by electrons}}{\text{number of electrons transferred}}. \quad (7)$$

Using the random-current approach as before,

$$\bar{E}_{12} = \frac{\bar{T}_{12} \int_{v_x}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{1}{2} m(v_x^2 + v_y^2 + v_z^2) \, dN,}{\bar{T}_{12} \int_{v_x}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dN} \quad (8)$$

where

$$dN = \frac{2m^3}{h^3} \frac{1}{1 + \exp(E - E_{m1}/kT_1)} v_x \, dv_x \, dv_y \, dv_z. \quad (9)$$

The term unity in equation (9) is negligible as before, and equation (8) is integrated, yielding

$$\bar{E}_{12} = E_0 + 2kT_1 \quad (10)$$

and

$$\bar{E}_{21} = E_0 + 2kT_2. \quad (11)$$

Substituting equations (6), (10) and (11) into equation (4), we obtain the final expression for heat transfer,

$$q_{e_{12}} = \frac{A_{a_{12}}}{A_n} \bar{T}_{12} C T_1^2 \exp\left[\frac{-(E_{w_2} - E_{w_s})}{kT_1}\right] 2k(T_1 - T_2). \quad (12)$$

According to quantum theory, not all the electrons which have sufficient energy to pass over a potential barrier will be transmitted through the potential barrier. The fraction passing through is given by $\bar{T}_{12}$, the transmissivity in equation (12). When the shape of the potential is assumed to be square, the transmissivity is given by a solution of the Schrodinger equation in one dimension [7].

$$\bar{T}_{12} = \left\{1 + \frac{E_0^2 \sin^2 l\sqrt{[\kappa(\bar{E} - E)_0]}}{4\bar{E}(\bar{E} - E_0)}\right\}^{-1}. \quad (13)$$

Now, consider an experiment such as Rogers'. Suppose that we hold the metal 1 side of the interface at a certain temperature T_A and the metal 2 side of the interface at a certain lower temperature T_B. Then, for this situation, equation (12) becomes

$$q_{e_{12}} = \frac{A_{a_{12}}}{A_n} \bar{T}_{12} C T_2^2 \exp\left[\frac{-(E_{w_1} - E_{w_s})}{kT_A}\right] 2k(T_A - T_B). \quad (14)$$

Now, suppose we reverse the interface temperatures. Heat will now flow from metal 2 to metal 1.

$$q_{e_{21}} = \frac{A_{a_{21}}}{A_n} \bar{T}_{21} C T_B^2 \exp\left[\frac{-(E_{w_1} - E_{w_s})}{kT_B}\right] 2k(T_A - T_B). \quad (15)$$

The ratio of these quantities is given by the expression,

$$\frac{q_{e_{12}}}{q_{e_{21}}} = \frac{A_{a_{12}}}{A_{a_{21}}} \frac{\bar{T}_{12}}{\bar{T}_{21}} \frac{T_A^2}{T_B^2} \exp\left[\frac{E_{w_1} - E_{w_s}}{k} \frac{T_A - T_B}{T_A T_B}\right]. \quad (16)$$

A consideration of the order of magnitude of terms in equation (16) clearly shows the presence of directional heat transfer. The ratio $\bar{T}_{12}/\bar{T}_{21}$ is approximately 1. The term T_A^2/T_B^2 is greater than unity, and, since $E_{w_1} > E_{w_s}$, the exponential term will be greater than 1. The term $A_{a_{12}}/A_{a_{21}}$ is usually unity, but may vary slightly from this

figure depending upon the hardness of the metals in contact. As Rogers points out [5] this effect is relatively unimportant. In any event, the exponential factor is controlling, and may be as great as 10, dominating the contribution of the ratio $A_{a_{12}}/A_{a_{21}}$.

DISCUSSION

This paper gives a qualitative explanation of asymmetric heat flow at the interface between dissimilar metals. Exact quantitative calculations of this effect cannot be made until we know more about the nature of the potential barriers existing between different metals at their surface of contact. If we assume a value of $E_{w_1} - E_{w_2} = 0{\cdot}3$ eV, and set $T_A = 350°\text{K}$, $T_B = 350°\text{K}$ as in Rogers' work, with $A_{a_{12}}/A_{n_{21}} = 1/150$, we obtain a value for $he_{12} - he_{21}$ of 150 chu/ft²h degC which compares favorably with Rogers' value of approximately 100 chu/ft²h degC.

In order to investigate the effect on asymmetric heat flow of using specimen metals with widely different thermoelectric potentials, Rogers conducted a series of experiments with the specimen pair T1 alloy (chromel)—T2 alloy (alumel). He found no directional effect with this pair. Since the widely different thermoelectric potentials produce a high potential barrier which virtually eliminates the electronic contribution to heat transfer, the only significant heat-transfer mechanism remaining is the phonon contribution. Thus, it can be concluded that the phonon heat-transfer mechanism is non-directional. Insertion of a mica shim between the stainless-steel–aluminum pair produced the same effect, eliminating asymmetric heat flow.

Williams [8] pointed out that the contact configuration of two metals might change with the direction of heat flow. As pointed out previously, this mechanism is not considered significant, but is taken into account in equation (16) by the factor $A_{a_{12}}/A_{a_{21}}$.

It can be shown that the tunneling effect is negligible, since the probability of transmission of electrons through the potential barrier is of the order of 10^{-10}, assuming a barrier thickness of 20 Å.

CONCLUSIONS

Additional experimental results are needed to test the theory developed in this paper. For instance, experiments could be performed over various temperatures and temperature differences, and values of $E_{w_1} - E_{w_2}$ could be back-calculated to test consistency, since this quantity varies only slightly with temperature. However, although solid-state theory qualitatively explains directional heat transfer, solid-state physicists will have to obtain accurate values of work functions of various metals and their oxide films and develop better theories for the contact potential before this effect can be predicted quantitatively.

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Résumé—Différents chercheurs ont trouvé que la résistance thermique de certaines interfaces métal–métal dépendait de la direction du flux de chaleur à travers ces interfaces. Cet article montre que ce phénomène peut s'expliquer par la théorie de la conduction thermique dans les solides.

Zusammenfassung—Versuche ergaben an den Berührungsflächen zweier verschiedener Metalle Kontaktwiderstände, die sich als abhängig von der Richtung des Wärmestroms erwiesen. Hier wird

gezeigt, dass ein derartiges Phänomen mit Hilfe der Theorie der Wärmeleitung in festen Körpern erklärbar ist.

Аннотация—Рядом исследователей было установлено, что на поверхности раздела при определенных сочетаниях металлметалл термосопротивление зависит от направления теплового потока через эти поверхности. В данной статье показано, что это явление можно объяснить на основе теории теплопроводности.