Two-Temperature Thermodynamic and Transport Properties of Hydrogen Plasmas

IEPC-2011-152

Presented at the 32nd International Electric Propulsion Conference,
Wiesbaden • Germany
September 11 – 15, 2011

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Abstract: The knowledge of two-temperature transport coefficients is of interest in the modelling of fluid flow and heat transfer processes in electric propulsion devices. Variations of species mole fraction, number density, specific enthalpy, specific heat at constant pressure, viscosity, electrical conductivity and thermal conductivity as a function of pressure, electron temperature and different degrees of temperature non-equilibrium are calculated for two-temperature hydrogen plasmas. To meet practical needs of modelling study of arcjet and MPD thrusters, pressure included in the calculation ranges from 0.0001 to 1 atmosphere, temperatures range from 300 K to 40000 K, and the ratio of electron temperature ($T_e$) to the heavy species temperature ($T_h$) ranges from 1 to 4. Results obtained for local thermodynamic equilibrium (LTE) under atmospheric pressure agree well with published results obtained for the same conditions.

Nomenclature

$b$ = impact parameter between interaction of species i and j
$c_p$ = specific heat at constant pressure
$D_{ij}$ = ordinary diffusion coefficient of species i and j
$D_{ij}^a$ = ambipolar diffusion coefficient of species i and j
$D_{ij}^b$ = binary diffusion coefficient of species i and j
$e$ = specific internal energy
$E_d$ = dissociation potential of hydrogen molecule
$E_i$ = ionization potential of hydrogen atom
$g_{ij}$ = degeneracy of j’th electronic excited level of species i
$h$ = Planck constant
$h_T$ = specific enthalpy
$i$ = species i
$j$ = species j
$k$ = thermal conductivity
$k_B$ = Boltzmann constant
$k_{e,r}$ = reactive thermal conductivity of electrons
$k_{h,r}$ = reactive thermal conductivity of heavy species

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During the past several decades, there has been increased interest in using numerical method to obtain useful physical insights of the electrical-thermal and electromagnetic plasma thrusters. 1 The plasma flow and heat transfer in arcjet and MPD thrusters are often characterized by the steep temperature, pressure, velocity, electrical potential gradients and the relatively short residence times. Experimental techniques provide much useful empirical data, but many quantities of interest are not accessible in the important regions of thrusters due to the limitation of small length scales and extremely hostile physical environment. And thus modelling plays an important role in understanding and predicting the physical behavior of such fluids inside realistic geometries of thrusters with different operating conditions.

The arcjet and MPD thrusters can be operated by a variety of different propellants such as hydrogen, argon, nitrogen, ammonia etc. Usually hydrogen is a favorable propellant since the hydrogen arcjet and MPD thrusters can achieve higher specific impulse. The coupled solution of mass, momentum and energy equations together with the electromagnetic field equations can be carried out once the thermodynamic and transport properties of the fluid investigation are known. There are some computations for thermodynamic and transport properties for a thermal hydrogen plasma available in the literature, while most of the reported results are for atmospheric pressure and include temperature ranges below 20000 K.2 There are increasing evidences show that maximum temperature inside constrictor of arcjet at high current (~100 A) may well exceed 30000 K.3 Moreover, the use of more and more sophisticated plasma diagnostics has shown that the assumption of local thermodynamic equilibrium (LTE) often fails in thermal plasmas, and the kinetic temperature of electrons, $T_e$, can be considered different from that of heavy species, $T_h$. This paper addresses this issue and calculated two-temperature thermodynamic and transport properties of hydrogen plasmas with pressure included ranges from 0.0001 to 1 atmosphere, electron temperature ranges from 300 K to 40000 K, and $\theta$ (= $T_e/T_h$) ranges from 1 to 4.

I. Introduction
II. Methods of calculation of thermodynamic properties of hydrogen plasmas

For a hydrogen atom, there is only one electron can be excited and released. So a two-temperature hydrogen plasma system consists of four species in our calculation, i.e., hydrogen molecules, H$_2$, hydrogen atoms, H, hydrogen ions, H$^+$, and electrons, e. When thermal non-equilibrium occurs, due to low energy exchange between heavy species and light electrons, a Maxwellian distribution of velocity characterized by the temperature $T_h$ is assumed for heavy species whereas the one for electrons is described by the temperature $T_e$. A parameter defined as $\theta = T_e / T_h$ is used to describe the thermal non-equilibrium degree. Number density of each species in the hydrogen plasma can be computed as a function of electron temperature and $\theta$ by solving the following set of equations.4

$$
n_i^2 = \exp\left(\frac{-E_i}{kT_e}\right) \left(\frac{\pi m_i k T_e}{\hbar^2}\right)^{3/2} \left(\frac{Q_{i,el}}{Q_{i,rot} Q_{i,vib}}\right)^{3/2}
$$

$$
n_i n_e = 2 \exp\left(\frac{-E_i}{kT_e}\right) \left(\frac{2\pi m_i k T_e}{\hbar^2}\right)^{1/2} \left(\frac{Q_{i,el}}{Q_{i,el}^0}\right)^{1/2}
$$

$$
p = k T_n + k T_e \sum_j n_j
$$

$$
n_i = n_i^0
$$

where the subscripts $d$, $a$, $s$ and $e$ are used to represent the hydrogen molecules, atoms, ions and electrons, respectively. The values of $E_d$ and $E_a$ for hydrogen plasma are 4.478 eV and 13.595 eV, respectively. $Q_{d,el}$, $Q_{d,rot}$, $Q_{d,vib}$, $Q_{a,el}$, $Q_{s,el}$, $Q_{s,rot}$, $Q_{s,vib}$ respectively represent the rotational and vibration partition function. The total partition function of a species ‘$i$’ is the product of its internal and translational partition functions and then the internal partition function is a product of rotational, vibrational, electronic and nuclear partition functions. For hydrogen atoms, hydrogen ions, the $Q_{i,rot}$ and $Q_{i,vib}$ are taken to be 1.0, and the contributions from nuclear partition functions are neglected in the formulation since the nuclei play no part in associated chemical processes. The rotational, vibrational partition function of molecules and total partition function of species $i$ are presented as follows.

$$
Q_{d,rot} = T / \theta_{rot}, \quad Q_{d,el} = \exp(-\frac{\theta_{rot}}{2T_d})[1 - \exp(-\frac{\theta_{rot}}{2T_d})], \quad Q_{s,el} = \sum_j \exp(-\frac{E_{i,j}}{k T_j})
$$

For hydrogen molecule, $\theta_{rot} = 87.5$ K and $\theta_{rot} = 6320$ K. 6 To meet the required accuracy of electronic partition functions, the maximum of variable $j$ is 15 in this calculation, and the data are taken from Ref. 5. For hydrogen molecules and hydrogen ions, $Q_{i,el}$ is taken to be 1.0.

The specific internal energy of the system including the dissociation and ionization potentials with volume $V$ is calculated from respective internal and translational partition functions as follows for the certainty of number densities.

$$
e = \frac{1}{\rho} \sum_i n_i k T_i \frac{\partial \ln Q_{i,rot}}{\partial T_i} + \frac{1}{\rho} \sum_i n_i k T_i \frac{\partial \ln Q_{i,el}}{\partial T_i} + \sum_i E_i / m_i
$$

where $E_i$ stands for the dissociation and ionization potentials for species $i$ from the ground state of hydrogen molecules. Thermodynamic properties under certain pressure such as specific enthalpy and specific heat at constant pressure are calculated from specific internal energy employing standard thermodynamic relationships as follows

$$
\rho = \sum_i n_i m_i, \quad h_e = e + p / \rho, \quad c_p = \sum_i \frac{\partial h_i}{\partial T_i}
$$

Equations (1-4) are solved together with Eq. (5) to obtain the number densities of the species present in hydrogen plasma, and then the density and specific enthalpy and specific heat are calculated according to Eq. (7).

III. Methods of calculation of transport properties of hydrogen plasmas

For calculating transport properties, the distribution function followed by the species in the plasma is assumed to be a first order perturbation to the Maxwellian distribution. The perturbation is then expressed in a series of Sonine polynomials,7 which finally through linearization of the Boltzmann equation leads to a system of linear equations that can be solved to obtain different transport coefficients.8 Whereas the elements in the matrix of the system of equations depend on the interaction between associated colliding species in terms of collision integrals, the number
of elements depends on the order of the chosen approximation. In the following we describe in brief the definition of collision integrals used for computing different transport properties from them.

A. Collision integrals

The collision integrals between species \( i \) and \( j \), \( \Omega_{ij}^{(s)} \), are defined as following\(^7\)

\[
\Omega_{ij}^{(s)} = \frac{2\pi k_T}{m_i^*} \int_0^\infty \int_0^\infty \exp(-\gamma_{ij}^\alpha) \gamma_{ij}^{2\alpha+1} (1 - \cos \chi) db \cdot dy_0
\]  
(8)

where \( m_i^* \) and \( T_0^* \) are defined as followings\(^7\)

\[
m_i^* = m_i m_j / (m_i + m_j), \quad T_0^* = (m_i T_i + m_j T_j) / (m_i + m_j)
\]  
(9)

In Eq. (8), \( \gamma_{ij}^\alpha \) is a function of the relative velocity \( g_0 \) between species \( i \) and \( j \) as

\[
\gamma_{ij}^\alpha = g_0 / \sqrt{2 k_T / m_i^*}
\]  
(10)

Usually, transport cross section are calculated either using the elastic collision cross section or from the numerical integrations of the interaction potential corresponding to the binary interaction considered, which then yield the angle of deflection, \( \chi \), defined as\(^7\)

\[
\chi = \pi - 2b \int dr / \left[ r^2 - 2V(r) / (m_i^* g_0^2 - b^2 / r^2) \right]
\]  
(11)

The collision integrals between charged species are calculated using screened Coulomb potentials according to Liboff.\(^9\) Other collision integrals between the species, \( \text{H}_2-\text{H}_2 \), \( \text{H}_2-\text{H} \), \( \text{H}_2-\text{H}^+ \), \( \text{H}_2-\text{e} \), \( \text{H}-\text{H} \), \( \text{H}-\text{H}^+ \), \( \text{H}-\text{e} \), are respectively given in Ref. 3 and Ref. 10. In order to cover the temperature range of our interest (300 K- 40000 K), available collision integral values are extrapolated based on the available data. For electron neutral interactions, it has been assumed that \( \Omega_{ij}^{(s)} = \Omega_{ij}^{(n)} \), which is suggested by Murphy.\(^11\) Recursion formula derived by Yun has been used for calculating \( \Omega_{ij}^{(s)} \) from \( \Omega_{ij}^{(n)} \) for the interactions between neutral and neutral.\(^12\)

B. Diffusion Coefficient

The binary diffusion coefficient between two species \( i \) and \( j \) for this diffusion model is defined as follow\(^7\)

\[
D_{ij} = 3k_T (T_i / (16 p T_i m_i^* \Omega_{ij}^{(s)}))
\]  
(12)

Once the species temperatures are known, sixteen binary diffusion coefficients associated with these cases are calculated using already available \( \Omega_{ij}^{(s)} \) values, and then the other diffusion coefficients derived from these binary diffusion coefficients can be computed employing standard relationships. The first order approximation to the ordinary diffusion coefficients can be written as an expression associated with matrix \( F \) and its cofactor \( F^\alpha \).\(^7\)

\[
D_{ij} = (F^\alpha - F^\beta) / (m_i^* F)
\]  
(13)

where the matrix \( F \) is defined as follow\(^7\)

\[
F_{ij} = 1 / \rho \left[ n_i D_{ij}^0 + \sum_{m=1}^n m D_{ij}^m \right] (1 - \delta_{ij})
\]  
(14)

The ambipolar diffusion coefficient is derived in terms of ordinary diffusion coefficients as\(^13\)

\[
D_{ij}^a = D_{ij} + \alpha \sum_i Z_i D_{ij} / \beta
\]  
(15)

where \( \alpha_i \) and \( \beta \) are expressions associated mass, number density, charge, ordinary diffusion coefficient and temperature of participating species as follows\(^13\)

\[
\alpha_i = \sum_j m_j n_j Z J_i / T_j, \quad \beta = \sum_i Z_i \sum_j m_i n_j Z J_i / T_j
\]  
(16)

C. Viscosity

The contribution to viscosity in hydrogen plasma comes mostly from heavy species, i.e., \( \text{H}_2 \), \( \text{H} \) and \( \text{H}^+ \), whereas the effect of electrons on viscosity are neglected due to the much smaller species mass. In this study, the first order approximation of viscosity is used.\(^14\)
The interested elements associated the matrix are calculated in terms of collision cross sections as

\[
\hat{Q}_{i,j}^{(1)}(\delta_{\eta} - \delta_{\nu}) m_j + 2 \nu m_j \hat{Q}_{i,j}^{(2,2)} (\delta_{\eta} + \delta_{\nu})
\]

(18)

D. Electrical Conductivity

Electrons play a determinant role in electrical conductivity property of plasmas. The third order approximation is employed as follows

\[
\sigma = \left( \frac{e^+ n_m n_i}{\rho k_{\text{e}} T_e} \right) \left( 3 n_i \rho \right) \sqrt{\frac{2 \pi k_{\text{e}}}{m}} \frac{q^{11}}{q^{21}} \frac{q^{12}}{q^{22}}
\]

(19)

\[q^{ij}\] is the element given by Devoto in terms of collision cross section \(\hat{Q}_{i,j}^{(\nu)}\).

E. Thermal Conductivity

In two-temperature hydrogen plasma system, the thermal conductivity is consisted of the translational thermal conductivity of electrons and heavy species, the reactive thermal conductivity of electrons and heavy species. The translational thermal conductivity of electrons and heavy species suggested by Devoto are shown as follows

\[
k_{\text{T}e} = \frac{75 n_i k_{\text{e}}}{8} \sqrt{\frac{2 \pi k_{\text{e}}}{m_e}} \frac{1}{q^{11} - (q^{12}) / q^{22}}
\]

(20)

\[
k_{\text{T}r} = \frac{-75 k_{\text{e}}}{8} \sqrt{\frac{2 \pi k_{\text{e}}}{m}} \frac{q^{10}}{q^{11}} \frac{q^{20}}{q^{21}} \frac{q^{10}}{q^{10}} \frac{q^{20}}{q^{20}}
\]

(21)

where \(q^{ij}\) is the element used for calculating electrical conductivity, and \(q^{ij}_{\nu}\) is a block which actually presents an array as following

\[
q_{ij}^{\nu} = \begin{bmatrix} q_{ij}^{11} & \cdots & q_{ij}^{1p} \\ \vdots & \ddots & \vdots \\ q_{ij}^{q1} & \cdots & q_{ij}^{pq} \end{bmatrix}
\]

(22)

\[|\eta|\] is the determinant formed from the numerator by deleting the last row and last column.

Based on the derivation presented in Ref. 13 and Ref. 16, if we neglect the influence due to the thermal diffusion and external force (except for internal electric field), the number flux vector of the species \(r\) in a multi-species control volume for a fixed total pressure can be written as

\[
\psi_r = \frac{n}{\rho k_{\text{e}} T_e} \sum_{j=1}^{n_m} T_j m_j D_{ij} \nabla p_j
\]

(23)

where \(D_{ij}\) is the ambipolar diffusion coefficient. The heat flux associated with the particle flux can be written as

\[
q_h = \sum_{r=1}^{n} \mathcal{H}_r \psi_r
\]

(24)
Inserting Eq. (23) into Eq. (24), the heat flux is expressed as

\[ q_r = -\left[ \sum_{j=1}^{m} \frac{n}{\rho k T_j} \sum_{i=1}^{n} T_i \frac{\partial f}{\partial T_i} \right] \nabla T - \left[ \sum_{j=1}^{m} \frac{n}{\rho k T_j} \sum_{i=1}^{n} T_i \frac{\partial f}{\partial T_i} \right] \nabla T_0 \]  

(25)

Then, the Eq. (24) can be rewritten as

\[ q_r = -k_{e,v} \nabla T_e - k_{h,v} \nabla T_h \]  

(26)

where

\[ k_{e,v} = \sum_{i=1}^{n} \Delta h_i \frac{n}{\rho k T_i} \sum_{j=1}^{m} T_j \frac{\partial f}{\partial T_j}, \quad k_{h,v} = \sum_{j=1}^{m} \Delta h_j \frac{n}{\rho k T_j} \sum_{i=1}^{n} T_i \frac{\partial f}{\partial T_i} \]  

(27)

where \( \Delta h_i \) is the heat of reaction and can be calculated as

\[ \Delta h_i = \frac{3}{2} k_e T_e + k_e T_e^2 \frac{\partial \ln Q_{\text{eqs},e}}{\partial T_e} + E_e - k_e \theta_{\text{e}} \left[ \frac{1}{2} + \exp\left( \frac{\theta_{\text{e}}}{T_e} \right) \right] \]  

(28)

\[ \Delta h_i = \frac{5}{2} k_e T_e + \frac{5}{2} k_e T_e^2 \frac{\partial \ln Q_{\text{eqs},e}}{\partial T_e} + E_e - k_e T_e \frac{\partial \ln Q_{\text{eqs},e}}{\partial T_e} \]  

(29)

Finally, the total thermal conductivity of two-temperature plasma system can be calculated as

\[ k = k_{e,v} + k_{h,v} + k_{e,v} + k_{h,v} + k_{e,v} \]  

(30)

IV. Results

In this section we present the variations of two-temperature thermodynamic and transport properties of hydrogen plasma with temperature, pressure and degrees of thermal non-equilibrium, \( \theta_e \), using the above methods of computation. The electron temperature varies from 300 K to 40 000 K and the pressure varies from 0.0001 atmosphere to 1 atmosphere.

A. Comparisons of plasma properties with those from previously reported results

Comparisons of calculated composition mole fractions, specific enthalpy, specific heat at constant pressure, viscosity and electrical conductivity of LTE hydrogen plasma under the case of \( \theta_e=1 \) with data available in the literatures are presented from Figs. 1 to 6, respectively. In these figures, the symbols of “BUAA” shows our results, “Miller” shows the results from Ref. 3, “Megli” shows results from Ref. 17 and “Murphy” shows the data provided by Prof. Murphy, respectively.

As showed in Fig. 1, dissociation of hydrogen starts around 1400 K. In hydrogen plasma system, a significantly rise in the partial pressure of the hydrogen atoms causes a decrease in the partial pressure of hydrogen molecules. With the further increase of temperature, the ionization of the hydrogen atoms starts at 10000 K. The mole fractions of hydrogen atom with different pressures are shown in Fig. 2, and our predict results show a good agreement with previous published results. Figures 3 and 4 respectively present the variations of the specific enthalpy and specific heat at constant pressure with the temperature at different pressures. In each curve of Fig. 4, the first peak
corresponds to dissociation of hydrogen molecules and the second one corresponds to ionization of hydrogen atoms, and the peaks are associated with the visible increase of specific enthalpy in Fig. 3. Figures 3 and 4 also show that our results of the thermodynamic properties obtained under local thermodynamic equilibrium are in good agreement with those already published in the literature.

B. Compositions

Figure 5 shows the variations of mole fractions of four species at atmospheric pressure as a function of electron temperature for different $\theta$ values. It is noted that the atom density shows an interesting behavior for larger $\theta$ values. As soon as a molecule is dissociated, the atoms will be immediately ionized because of the high electron temperature. As a consequence, the atom density decreases with the increase of $\theta$, and the similar behavior of oxygen density variation with $\theta$ in nonequilibrium plasma system also shown in Ref. 18.

Figure 5. Variations of mole fractions of different species for hydrogen plasma in two-temperature state with electron temperature and different $\theta$ values at 1 atm: (a) $\theta=1$, (b) $\theta=2$, (c) $\theta=3$, (d) $\theta=4$.
Variations of the electron number density as a function of electron temperature for different $\theta$ values and pressures are presented in Fig. 6. It can be noted that the electron number density increases with the increase of $\theta$ at the same pressure. The peaks in electron number density occur around $T_e$ equal to 17000 K at atmospheric pressure, and the peaks shift to lower values of $T_e$ as the pressure falls down to 0.01 atmospheric pressure, because the change of equilibrium state of ionization-recombination reactions also depend on the pressure. In a summation, dissociation of molecules serves as the root cause for the generation of every other species present in the system.

![Figure 6](image1.png)

**Figure 6.** Variation of electron number densities for hydrogen plasma in two-temperature state with electron temperature: (a) $p=1$ atm, (b) $p=0.01$ atm.

### C. Thermodynamic properties

![Figure 7](image2.png)

**Figure 7.** Variation of specific enthalpy for hydrogen plasma in two-temperature state with electron temperature: (a) $p=1$ atm, (b) $p=0.01$ atm.

![Figure 8](image3.png)

**Figure 8.** Variation of specific heat at constant pressure for hydrogen plasma in two-temperature state with electron temperature: (a) $p=1$ atm, (b) $p=0.01$ atm.
The variations of specific enthalpy are presented in Fig. 7 as a function of electron temperature for various values of θ and different pressures. It is observed that specific enthalpy is almost linear with $T_e$ unless some reactions like dissociation and ionization occur during certain temperature ranges. Similar to earlier observations, the behavior for higher θ are distinctly different from behavior at lower θ caused by low number density and temperature of heavy species. Except a drift of the curves toward higher $T_e$, the overall variation in enthalpy with increasing of pressure is small. Specific heat at constant pressure ($c_p$) for electrons and heavy species are derived from the respective enthalpy component behavior with respect to $T_e$ and $T_{th}$, and $c_p$ of the system of two-temperature hydrogen plasmas is presented in Fig. 8. It is observed that the height of the peaks observed is due to the release of dissociation and ionization potential in the system.

D. Transport properties

![Figure 9. Variation of viscosity for hydrogen plasma in two-temperature state with electron temperature: (a) $p=1$ atm, (b) $p=0.01$ atm.](image)

Figure 9 presents the variations of viscosity with electron temperature, θ and pressure. As is known, the momentum transport of heavy species in gas is mostly responsible for viscosity of the system. It is seen that, below 8000 K, the viscosity is dominated by the neutral-neutral interactions and the viscosity increase almost linearly with the increase of the temperature. Once ionization starts, long-range coulomb interaction increases and results in the observed drop in the viscosity value. With increasing θ, the peaks in the viscosity shift towards higher $T_e$ values. The viscosity also increases with an increasing pressure and the peaks shift towards higher $T_e$ values.

![Figure 10. Variation of electrical conductivity for hydrogen plasma in two-temperature state with electron temperature: (a) $p=1$ atm, (b) $p=0.01$ atm.](image)

Figure 10 presents the variations of electrical conductivity with electron temperature, θ and pressure. Electrical conductivity is a property of electrons alone due to the much higher velocity. For higher pressures, it exhibits higher values due to higher number densities of electrons. With higher θ values, a visible increasing is found once the ionization temperature has been reached, and then quickly stabilizes near the values observed for lower values of θ at that $T_e$. 

The 32nd International Electric Propulsion Conference, Wiesbaden, Germany
September 11 – 15, 2011
Figures 11, 12 and 13 respectively present the translational thermal conductivity of heavy species, reactive thermal conductivity and total thermal conductivity of system for various $\theta$ values and pressures. The reactive thermal conductivity makes a great contribution to total thermal conductivity during the temperature range that dissociation or ionization reaction occurs, and it is associated with the variation of the number density of species which basically depend on their temperatures. For a higher $\theta$ value, the peaks of reactive thermal conductivity associated with dissociation and ionization reactions shift towards each other, achieve a new higher peak. At low
temperature, the translation contribution of heavy species is dominant. After the ionization, when $T_e$ increases, the electron translational thermal conductivity increases, and dominates the behavior of the thermal conductivity, especially as $\theta$ increase.

V. Conclusion

Compositions, thermodynamic and transport properties are calculated in this paper for a two-temperature hydrogen plasma system under local chemical equilibrium conditions. It is assumed that electron temperature is different from that of heavy species which are constituted of hydrogen molecules, hydrogen atoms, and hydrogen ions for a plasma system. The property values are given for a pressure ranges from 0.0001 to 1 atm, and for a range of electron temperatures from 300 K to 40 000 K. This range is wider than the ranges usually found in the literature because it was found necessary to provide values for numerical simulation calculations of arcjet and MPD thruster. The two-temperature considered is expressed as the ratio $\theta$ of electron temperature to heavy particle temperature and the non-equilibrium parameter of $\theta$ varies from 1 to 4. It has been shown that the $\theta$ has a strong influence on all the properties over a wide range of temperatures. The behavior of thermodynamic and transport properties of hydrogen plasma depending on electron temperature, pressure and $\theta$ are discussed. Results of calculations for $\theta = 1$ represent LTE state values of the properties, and the data obtained for these conditions compare favorably with those published previously.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (Nos. 11072020, 50836007).

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