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MECHANISM OF MICROWAVE HEATING OF DIELECTRIC AND MAGNETIC MATERIALS BY MEANS OF ATOMISTIC THEORIES

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Abstract

Heating of materials by microwaves is selective and dependent on their properties. Dielectric materials are efficiently heated by the electric field of microwaves while magnetic materials by the magnetic field. We emphasize that electron system first responds to microwaves to absorb energy, and that a path of steady and irreversible energy flow from electrons to lattice atoms must be present. We showed using a classical Heisenberg model that dissipation is essential for energy relaxation [Phys.Rev.B, 2009]. However, the origin of dissipation has yet to be identified where mediation of electromagnetic waves - spin waves and/or hypersound waves is highly expected. We are performing both classical Heisenberg and ab initio simulations to elucidate the origin of dissipation from atomistic level. Our concern is whether such dissipation is materials specific or universal. In this paper, we will present our theoretical study of magnetic material heating in the microwave magnetic field.

Introduction

Selectivity and high efficiency of heating and high yield of chemical reactions in polar and organic molecules are well-known aspects of microwave materials processing. As the most ordinary dielectric liquid - water H_2O, we have shown its heating mechanism under microwaves due to rotational response of water molecules and subsequent energy relaxation by molecular collisions by using Newtonian molecular dynamics [1]. Heating was enhanced even when small amount of salt ions was added to water or at elevated temperature, both due to weakening of hydrogen-bonded network of water [2].

Then, we dealt with the heating of solid materials by microwaves, including dielectric materials [3-5] and magnetic materials [6]. In the former we performed both theoretical analysis of Mie theory and numerical simulation solving the Maxwell equations under the given dielectric constants. When solid material consists of powders of μm (micron meter) grain sizes, the electromagnetic waves penetrate through the sample for a long distance, typically of the order of centimeters with realistic dielectric constants. We showed that the optimum heating is realized for the grain size of twice the skin depth, typically $3\mu m$ for copper particles [3].

As for the heating of magnetic materials, we showed for magnetite that the magnetic energy of microwaves agitates the electron spins which occurs off-resonantly [6]. The energy first absorbed by electron spins is then transferred to lattice atoms by relaxation process. The maximum heating of magnetite at the Curie temperature 858K and its continuation to higher temperatures, and the optimal heating at 2GHz microwave, were theoretically obtained. These features agreed very well with experimental evidences.

However, in the above study of magnetite heating, the relaxation – dissipation was taken into account as the relaxation time constant, which was assumed to be in the nano sec range. The purpose of the present paper is to show our current progress in the energy transfer from electrons to lattice atoms.

Heating of Magnetite by Microwave Magnetic Field

Absorption of magnetic energy of microwaves by magnetic material was studied for magnetite [6]. Electrons are localized in insulator magnets like magnetite, and the Heisenberg model for electron spins was a proper tool of the study. The Heisenberg spin equation was time integrated under the applied microwave, and three things were obtained theoretically. (1) The heating occurs due to the response of magnetization to microwaves, which originates from electron spins residing in the unfilled 3*d* shell. Their nonresonant response causes a large change in the internal energy through the exchange interaction between spins, which is by a factor of $J_{AB}S / g\mu_B B\,(\gg 1)$ larger than the Zeeman term. (2) The heating persists above the Curie temperature T_c because each electron spin is able to respond to the alternating magnetic field of microwaves even above T_c. This energy change will then be dissipated to lattices and contribute to heating. (3) Hematite Fe_2O_3 which has only weak spontaneous magnetization shows much less response to microwaves than magnetite. (4) The heating of titanium oxide having oxygen defects TiO_{2-x} ($x>0$) by the microwave magnetic field is explained by our theory in terms of intrinsic (spontaneous) magnetization.

Energy Transfer from Microwaves to Lattice Atoms

As described in the previous section, we have proven that the magnetic energy of microwaves is first absorbed by electron spins residing in the 3d atomic orbital when magnetite is irradiated by microwaves [6]. In order to complete the study of microwave energy absorption by magnetic materials, it is necessary to find the energy relaxation path from the electron spins to lattice atoms.

Here, we describe our theoretical study of the energy relaxation from spins to atoms within the classical theory. We adopt the Heisenberg model for electrons spins, and the Newtonian dynamics for nuclei. We denote the i-th atomic position $\mathbf{r}_i(t)$ at time t, its electron spin $\mathbf{s}_i(t)$, and the microwave magnetic field $\mathbf{H}(t)$. Then, the Hamiltonian for the electron spins is given by

$$H_e = -\sum_{i,j} J_{ij}(\mathbf{r}_i - \mathbf{r}_j)\mathbf{s}_i \bullet \mathbf{s}_j + g\mu_B \sum_i \mathbf{s}_i \bullet \mathbf{H} \qquad (1)$$

where we introduce the spatial dependence of exchange interactions J_{ij} which arises from overlapping of two adjacent atoms. The exchange interaction coefficients can be determined by ab initio (quantum mechanical) simulation techniques. The time change of the spins is calculated by the Heisenberg spin equations,

$$\hbar \frac{d\mathbf{s}_i}{dt} = 2\sum_{i,j} J_{ij}(\mathbf{r}_i - \mathbf{r}_j)\mathbf{s}_i \times \mathbf{s}_j - g\mu_B \sum_i \mathbf{s}_i \times \mathbf{H}(t) \qquad (2)$$

To deduce the forces on atomic nuclei, we note the phenomenon that the magnetic material is distorted along the magnetic axis when it is pressed by external force. Thus, it is natural to assume that atoms respond to the change in the electron Hamiltonian such that it is preserved, which in turn exerts forces on atoms. The force on nuclei at the position $\mathbf{r}_i$ may be given by

$$\mathbf{F}_i^{(s)}(\mathbf{r}_i) = -\frac{\partial H_e}{\partial \mathbf{r}_i} = \sum_j \frac{dJ_{ij}(r_{ij})}{dr} \frac{\mathbf{r}_i - \mathbf{r}_j}{|\mathbf{r}_i - \mathbf{r}_j|} \mathbf{s}_i \bullet \mathbf{s}_j \qquad (3)$$

where $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ is the distance between the i-th and j-th atoms. Also, a restoring forces on lattice atoms in magnetite arises from the Coulomb (electrostatic) interactions,

$$\mathbf{F}_i^{(c)}(\mathbf{r}_i) = \sum_j \frac{q_i q_j}{|\mathbf{r}_i - \mathbf{r}_j|^2} \frac{\mathbf{r}_i - \mathbf{r}_j}{|\mathbf{r}_i - \mathbf{r}_j|} \qquad (4)$$

since electrons in magnetite are localized and it consists of ionic bonds of Fe^{2+}, Fe^{3+} and O^{2-} ions. The equation of motion for nuclei is,

$$m_i \frac{d\mathbf{v}_i}{dt} = \mathbf{F}_i^{(s)}(\mathbf{r}_i) + \mathbf{F}_i^{(c)}(\mathbf{r}_i), \quad \frac{d\mathbf{r}_i}{dt} = \mathbf{v}_\mathbf{i} \qquad (5)$$

Through eqs.(2)-(5), the microwave magnetic field couples nonlinearly with electron spins and nuclei in which spins are affected also by atomic motion through eq.(2).

Currently, we are performing above numerical simulations for magnetite $FeO{\bullet}Fe_2O_3$ for which the Heisenberg model is safely used due to localization of electrons on atoms. The sample magnetite consists of 3x3x3 fundamental domains, each of which includes A and B sub-lattices with 24 irons of Fe^{2+} or Fe^{3+} and 32 oxygen O^{2-} atoms. We first do simulations without microwaves to determine the background waves in the equilibrium state. The Coulomb forces serve as restoring forces to maintain lattice equilibrium, which however give rise to rapid atomic vibrations in THz range. Then, we will apply microwave magnetic field and find the waves, possible candidates are those including spin waves and acoustic waves [7].

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