## §14. Materials Research and Development by the First-Principle DFT Molecular Dynamics Simulations

Tanaka, M., Ignatenko, M. (Chubu Univ.),
Kono, H. (Tohoku Univ.), Maruyama, K. (RIKEN)

Magnetic metal particles (Fe, Co, Ni) and their oxides (magnetite, $TiO_{2-x}$) are efficiently heated by the magnetic field component of microwaves [1,2]. We showed the heating mechanism of metal oxides by the Heisenberg model [3], on top of the heating of metal powders by microwaves [4]. Namely, the electron spins of metal oxides in the 3d unfilled shell, which are the source of spontaneous magnetization, are responsible for the heating. The exchange interactions of spins result in the energy absorption much larger than that expected through the Zeeman term [3]. The heating of magnetite starting at room temperature persists much above the Curie temperature, and becomes maximal around 2 GHz (Fig.1). This reveals the nonresonant nature of the microwave heating. Our theory based on 3d electron spins, i.e. spontaneous magnetization, naturally explains that titanium oxides are heated only when oxygen defects are present $TiO_{2-x}$ (x>0).

We describe our progress in the quantum mechanical calculations of microwave heating of metal oxide and metal particles having spontaneous magnetization [5]. In the former material, we try to obtain the exchange interaction coefficients $J_{ij}$ which is to be used in the Heisenberg model. This is evaluated by the DFT (density functional theory) quantum mechanical code. In the latter for which electrons are itinerant, we use the TDDFT (time dependent DFT) code and calculate the linear response of electrons to obtain the imaginary part of magnetic susceptibility Im $\chi$. The time-dependent Kohn-Sham equation is

$$i\hbar\frac{\partial}{\partial t}\varphi_i(r,t)=\left[-\frac{\hbar^2}{2m}\nabla^2+\upsilon_{KS}(r,t)\right]\varphi_i(r,t),$$

where the KS potential is represented by

$$\upsilon_{KS}(r,t)=\upsilon_{ext}(r,t)+\int d^3r'\frac{n(r,t)}{|r-r'|}+\upsilon_{xc}(r,t).$$

In the DFT, the electron density is given by

$n(r,t)=\sum_i^{occ}|\varphi_i(r,t)|^2$, where the subscript $i$ in the summation denotes electrons.

For the TDDFT simulations on the Hitachi SR Supercomputer at NIFS, we have worked hard to port the Octopus code [6]. Since the code was developed on Linux machines – the world standard of supercomputing, subtle differences between the Linux and SR Fortran compilers made great difficulties, besides a bug in the FORALL statement on the SR compiler. Finally, we have resolved the problem which is found to originate from the semantics difference of two compilers in the save attributes of subroutine return values. Currently, we are calculating the equilibrium state of magnetite using the steady state first principle DFT code.

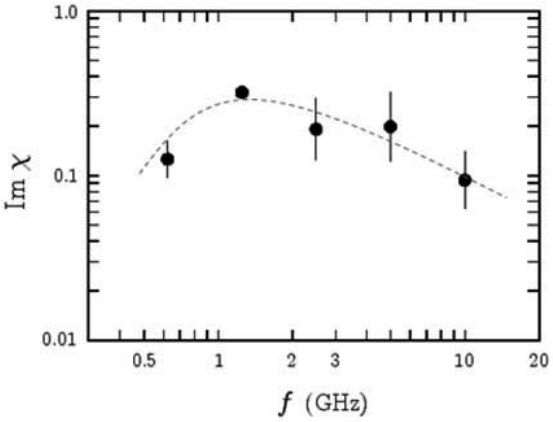


Fig1. The dependence of the imaginary part of magnetic susceptibility against the microwave frequency. This agrees well with experiments.

1. R. Roy, D. Agrawal, J. Cheng and S. Gedevanishvili, Nature 399, 668 (1999).
2. M. Sato, A. Matsubara, K. Kawahata, O. Motojima, T.Hayashi and S.Takayama, pp.277-280, *Microwave and High Frequency Heating* (AMPERE-10, Italy, 2005).
3. M. Tanaka, H. Kono, and K. Maruyama, Phys. Rev. B, 79, 104420 (2009).
4. M.Ignatenko, M.Tanaka and M.Sato, Jpn.J.Appl. Phys., 48, 067001 (2009).
5. C.Kittel, *Introduction to Solid State Physics*, Chap. 14-16, 6th edition (Wiley, 1986).
6. A.Castro et al., *Time Dependent DFT code – Octopus*, http://www.tddft.org/.
7. *Time-Dependent Density Functional Theory*, edited by M.Marque et al. (Springer, 2006).