Polarized Infrared Reflectance Spectra of a Series of Triangular Antiferromagnets, Pd(dmit)$_2$ Salts: Experimental Estimation of Electronic Anisotropies

Masafumi Tamura[1,2], Takashi Yamamoto[1,3], Reizo Kato[1], and Kyuya Yakushi[4]
[1]*Condensed Molecular Materials Lab., RIKEN, Japan*
[2]*Department of Physics, Faculty of Science and Technology, Tokyo University of Science, Japan*
[3]*Department of Chemistry, Faculty of Science, Osaka University, Japan*
[4]*Institute for Molecular Science, Japan.*
Email: qra_tam@ph.noda.tus.ac.jp

The anion radical salts of a metal complex Pd(dmit)$_2$ provide a rich variety of unconventional physics arising from the frustrated quantum spins on their triangular lattice structures formed of the dimers, $[Pd(dmit)_2]_2^-$. The low-temperature magnetic behavior varies with the counter cations [1-4], which is believed to be driven by the change in in-plane anisotropy of the interdimer exchange couplings (J) or transfer integrals (t). Therefore, experimental information on the parameters, t or J, is desired. We have measured polarized infrared reflectance spectra of the Me_4P, Et_2Me_2P, $EtMe_3As$ salts (AF ground states), the Et_2Me_2Sb and monoclinic $EtMe_3P$ salts (gapped ground states), and the $EtMe_3Sb$ salt (spin-liquid ground state) at various temperatures. Figure 1 shows the reflectivity data of the monoclinic $EtMe_3P$ salt at 10 K. The spectra indicate another characteristic of the Pd(dmit)$_2$ salts. Owing to the strong dimerization, the intradimer optical transition of these salts appears at ca. 10,000 cm $^{-1}$, which is well separated from the interdimer charge transfer (CT) transition across the Mott gap observed in the mid-infrared range. Thus, the optical conductivity data corresponding to the CT transitions obtained via the Kramers-Kronig analyses, give direct estimation of the interdimer t's, when combined with structural data. We will report on the systematic variation of the experimentally estimated interdimer t's with cations, and frustration-driven change in the Mott gap, for the first time in the Pd(dmit)$_2$ salts.

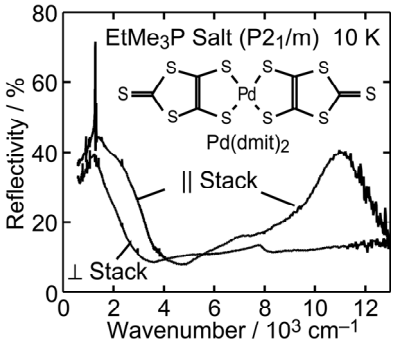

Fig. 1. Polarized reflectance spectra of the $EtMe_3P$ salt at 10 K.

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