

Resonance Effects on Two-Photon Photoemission of Lead Phthalocyanine Thin Films on Graphite

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Two-photon photoemission (2PPE) spectroscopy has been performed for lead phthalocyanine films on Highly Oriented Pyrolytic Graphite (HOPG) surface. Fully resolved occupied and unoccupied levels and resonant optical transitions between them are characteristics of the film surface [1]. 2PPE peaks due to photoemission from normally unoccupied levels (1ω peaks) and those due to coherent two-photon process from occupied levels (2ω peaks) show unexpected variations in intensities and widths when the pump photon energy crosses the resonances[2]. At around a resonance between molecule-derived levels, we find an intensity switching in which only the 2ω peak appears at photon energy below the resonance, and at above the resonance, the 1ω peak becomes prominent and the 2ω peak becomes very weak. The 1ω peak is broadened with increasing photon energy. These results cannot be interpreted by a simple energy level scheme, and point to further understanding of 2PPE process. The broadening of the 1ω peak can be interpreted in the framework of the hole scattering mechanism[3], and suggests that the resonantly excited state is not a molecular exciton in which the hole and electron are localized within a molecule.

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[2] M. Shibuta, K. Yamamoto, K. Miyakubo, T. Yamada, T. Munakata, *Phys. Rev. B.* **81**, 115426 (2010).

[3] M. Sakaue, T. Munakata, H. Kasai, A. Okiji, *Phys. Rev. B* **68**, 205421 (2003).