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AN ESR IMAGE OF NITRODIOXIDE MOLECULES ADSORBED ON A COPPER ROD AT 10 K

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Spatial distribution of paramagentic nitrodioxide molecules adsorbed on a rod-like cryo-tip sample holder, was measured using ESR imaging. This technique will serve to investigate surface diffusion of reactant species on catalyses.

Surface diffusion like lateral motion of an adsorbate along a metal surface, gives information on the potential experienced by the adsorbate as it moves. It occurs at annealing and order-disorder transformations and may be rate limiting steps in desorptions of dissociated species in catalytic processes. The reactive adsorbate walks randomly above a particular temperature and collides with different species. Some of them react with each other to form molecules.

In order to develop a technique available for the investigation of surface diffusion, ESR imaging was applied to an experiment on paramagnetic nitrodioxide (NO_2) molecules adsorbed on a rod-like copper cryo-tip cooled at about 10 K. ESR imaging which represents spatial distribution of paramagnetic species, has recently well developed and been applied to various fields of science.[1-9] Figure 1 illustrates the scheme of a cryogenic system with an ESR cavity. The apparatus for ESR imaging and its procedure were described in the previous paper.[4] A closed cycle refrigerator (Displex CS-202) and a vacuum shroud (WMX-1A/15) were used to carry out the cryogenic research. NO_2 gas was introduced and adsorbed on the cryo-tip in the direction marked with a thick arrow. After adsorption, the cryo-tip was moved down into the quartz tube and inserted into the microwave resonant

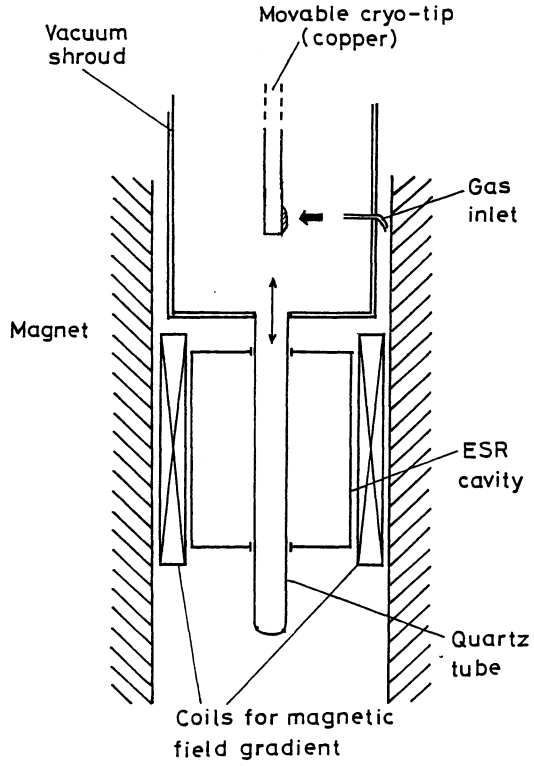

Fig. 1. A sketch of the cryogenic system for ESR imaging of adsorbed NO_2 molecules.

cavity with two coil pairs attached on its both oustsides to generate $\partial H_z/\partial x$ and $\partial H_z/\partial z$. Once the cryo-tip is fixed in the cavity it can not be rotated at all, so that merely the rotary magnetic field gradient method is available to acquire 36 projection spectra nesessary for 2-D image reconstruction.[4)]

A typical amorphous ESR spectrum of NO_2 molecule was observed without magnetic field gradient as in Fig. 2(a), indicating that the complex response function is due to the superposition of resonance lines with different linewidths and seems one of the most severe patterns for deconvolution. Nevertheless the m_I= -1 line could be used as a response function and a convoluted spectrum in Fig.2(b) was measured with 1.00 T/m of resultant field gradient from $\partial H_z/\partial z$ and $\partial H_z/\partial x$. Its left tail was determined by making reference to that of the lowest magnetic field line. Figure 3 represents the concentration of NO_2 molecules adsorbed on the surface of the rod-like sample holder (diameter 3 mm). The intense signal is located at the site where NO_2 gas was introduced and deposited. Surface diffusion phenomena and chemical reactions will be visualized from the ESR images after heating up above a particular temperature.

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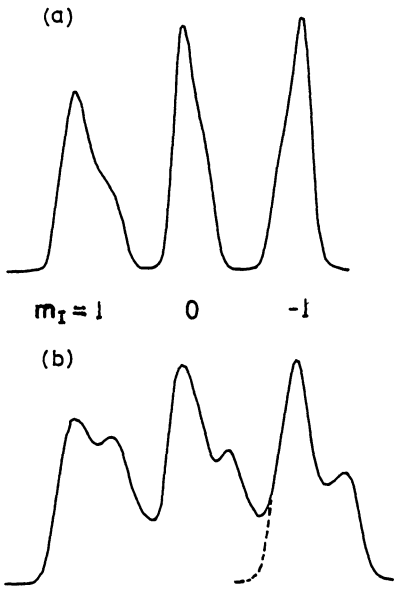

Fig. 2. ESR adsorption spectra of NO_2 at 10 K; (a) without magnetic field gradient, and (b) with it.

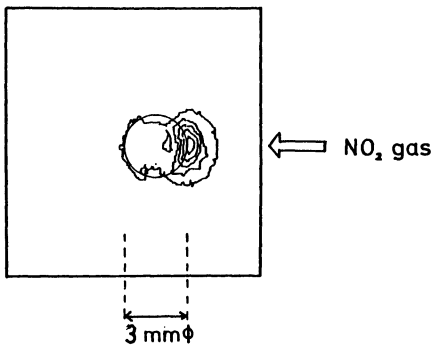

Fig. 3. A contour line map of NO_2 adsorbed on a copper cryo-tip.

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