MAGNETIC SUSCEPTIBILITY AS A PROBE OF OXYGEN SYSTEMS OF RESTRICTED GEOMETRY

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JOURNAL DE PHYSIQUE *Colloque C6, supplément au n° 8, Tome 39, août 1978, page* C6-334

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Résumé.- Nous discutons les résultats obtenus sur la susceptibilité magnétique des phases d'oxygène adsorbé sur deux substrats. Nous observons que la géométrie particulière aux films d'oxygène et que les propriétés microscopiques des substrats influencent le comportement de cette susceptibilité.

Abstract.- Experiments are discussed in which the magnetic susceptibility of adsorbed phases of oxygen has been studied. The observed behavior is seen to be influenced both by the restricted geometry of the oxygen films and by microscopic properties of the substrates.

Recent papers /1,2/ have reported some measurements of the magnetic susceptibility of oxygen physisorbed on two substrates, Vycor and Grafoil. In many investigations, the substrate serves primarily to define a restrected geometry. However, the substrate cannot usually be considered as a smooth plane which presents a single Van der Waals potential to an adsorbent. In general, the substrate potential and microscopic geometry will play an evident role in the physics of adsorbed systems. I shall briefly examine adsorbed oxygen experiments for indications of how one might select substrates to create some interesting physical conditions.

Grafoil does not appear to register any phase of adsorbed oxygen /3/ and might therefore provide a relatively straightforward enforcer of two-dimensional geometry for this system. (The interaction between the axis of the molecule and the spin is weak. Therefore, one might expect the effective spin dimensionality to be 3 for temperatures greater than those of liquid helium even if the molecules are oriented perpendicular to the substrate either by the substrate-molecule interaction or by the requirements of packing in the various surface phases.)

The substrate Vycor provides a very interesting contrast to Grafoil. Whereas both substrates have large ($\sim 10^3$ K) binding energies for typical adsorbents, the (periodic) variation across the graphite surface is $\sim$ 10 K. Vycor, on the other hand presents a distribution of binding energies with $\sim 10^2 - 10^3$ K and it is supposed that the existence of his wide distribution enforces a random localization of molecules on the substrate surface.

Susceptibility data for submonolayer coverages of oxygen on Grafoil and Vycor are shown in the figure.

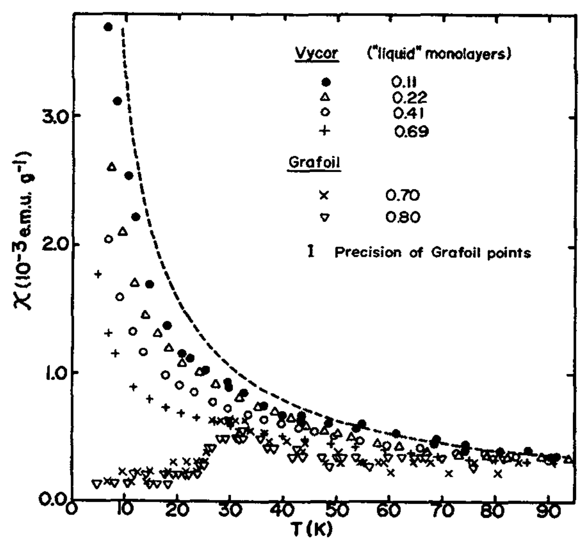

Fig. : Magnetic susceptibility of sub-monolayer oxygen adsorbed on Vycor and Grafoil substrates. Coverages are expressed as a fraction of a "liquid" monolayer - a monolayer of molecules packed with mean area of occupation 12.9 Å^2 per molecule. (The precision of the Vycor data is not indicated as it is about an order of magnitude better than that of the Grafoil data.) The dashed line indicates the Curie behavior appropriate to oxygen spins.

The Grafoil data indicate a transition in the region 25 - 30 K for both averages. Although enough coverages have not yet been studied, it is anticipated that a fairly constant and large value of paramagnetic Curie temperature will characterize all but the smallest sub-monolayers. This is because, on Grafoil, oxygen molecules adsorbed at several tens of degrees absolute should form "islands" in which the local environment of a molecule is fairly unchanging. Thus, the effective coverage is constant - only the total amount of oxygen adsorbed varies. In the case of oxygen on Vycor, at low coverages the data tend toward Curie-like behavior appropriate to a system of non-interacting spins. With increasing coverage they become more characteristic of an amorphous antiferromagnetic system, with increasingly negative paramagnetic Curie temperatures and excess low-temperature susceptibility.

It appears that the first monolayer on Vycor is constrained to be amorphous by the substrate forces. Interesting hysteresis effects in the susceptibility dependence on temperature are observed with coverages greater than one monolayer /1/ which might be explicable in terms of competition between ordering tendencies in upper layers and the requirements of the interface with the disordered first layer. The absence of these particular effects in the oxygen-Grafoil system is, therefore, not unexpected. Further experiments are planned to investigate the development of the hysteretic behavior at higher coverages using porous glass with larger pore sizes.

This work is supported by the Department of Energy through Contract No. EY-76-S-02-3151, Technical Report No. COO-3151-82.

References

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