

Observation of ferromagnetic properties of a monolayer of oxygen on the surface of gold

V. D. Borman, B. I. Buttsev, V. A. Konakov, B. I. Nikolaev, and V. I. Troyan

Moscow Engineering Physics Institute

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Investigations of the thermomagnetic effect (TME) in a Knudsen gas at temperatures 77–150 K have revealed that a monolayer of oxygen on a surface of gold has ferromagnetic properties.

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It is known that the study of magnetic phenomena in two-dimensional systems is of fundamental importance in solid-state physics and in the physics of phase transitions. It is therefore of interest to consider as objects of this type adsorbed monolayer coatings on solid surfaces. There are at present, however, no reported investigations

that might stimulate research in this field. In the course of work on extending the possibility of the TME^{11,21} for the investigation of surfaces, we have observed that the magnetic field influences the state of a chemisorbed oxygen layer on the surface of polycrystalline gold. In particular, we observed characteristic attributes of ferromagnetism, such as a dependence on the prior history of the magnetic action, saturation, and hysteresis of the states.

The experiments were performed with N₂ gas interacting with the surface of gold at temperatures 77–400 K. The “hot” surface ($T=130\text{--}400$ K) was a mica plate on which gold was sputtered (sputtering thickness $\sim 1 \times 10^{-5}$ cm, $R \approx 50 \Omega$), and the “cold” surface ($T=77\text{--}300$ K) was a layer of Au sputtered on a brass plate. These plates were separated by a distance $L=0.2$ cm in a vacuum chamber cooled by liquid nitrogen and placed in the gap of an electromagnet. The gold layer on the mica surface acted as a thermal resistor connected in a measuring bridge. We measured the change of the resistance of this layer in response to the change in a magnetic field with an energy flux Q in the N₂ gas, both as a result of precession of the molecules and as a result of the change in the elastic scattering of the N₂ molecules by the Au when the field acted on the state of this surface. The N₂ pressure was chosen to be $P \approx 1 \times 10^{-3}$ Torr in such a way that the mean free path of the molecules was $\lambda \geq 10L$. The observation of the TME is possible when $\omega\tau \sim 1$ [$\omega = \gamma H$, $\tau = L/\bar{v}$, and for N₂ we have $\gamma \approx 1.25 \times 10^3$ (sec Oe)⁻¹ and $H \sim 100$ Oe]. Prior to the experiments the surfaces were conditioned by heating in an atmosphere of oxygen which, in contrast to nitrogen, is chemisorbed on the gold¹³ (the absorption energy is $E_a \approx 4\text{--}5$ eV, the lifetime on the surface is $t = t_0 e^{E_a/T}$, where $t_0 \sim 10^{-13}$ sec is much longer than the time of the experiment). Under these conditions the N₂ molecules were scattered by the Au surface coated with the monolayer of chemisorbed oxygen. The systematic reproducibility of the results for different surface samples indicated that the concentration of the possible impurities in the surface layer was apparently small. The absence of parasitic phenomena in the measurements was confirmed by the zero unbalance of the bridge when the field was turned on in the case when the vacuum chamber was filled with the inert gas Ar (or when it was evacuated to $P \leq 10^{-6}$ Torr).

The results of the experiments are shown in Fig. 1. The plots of $\Delta Q(+H)$ (curves 1, 2, and 3) were obtained in a field $\mathbf{H} \parallel \mathbf{k}$ (\mathbf{k} is the normal to the surface) after power application, for a short time (~ 10 sec) after application of fields $H_0 = 3.5$ kOe, -3.5 kOe, as well as $H_0 = 0$, respectively ($\mathbf{H}_0 \perp \mathbf{k}$). It is seen that $\Delta Q > 0$, and when the sign of \mathbf{H}_0 is reversed the plots of $\Delta Q(H)$ differ from one another. If $H_0 = 3.5$ kOe, then when the field is increased, $\Delta Q(H)$ (curve 1) reaches a maximum at $H \approx 60$ Oe and tends to a value ΔQ_{sat} as $H \rightarrow \infty$. This corresponds to a precession mechanism of the variation of Q , when the polarized gas molecules interact with a surface that has not been altered in the field H . The maximum at $\omega\tau \sim 1$ corresponds to the predictions of the theory of the TME.^{11,21} As $\omega\tau \rightarrow \infty$, the orientations of the moments of the molecules M in the plane $\perp \mathbf{H}$ are completely averaged out and $\Delta Q \rightarrow \Delta Q_{\text{sat}}$. Inasmuch as $\omega\tau > 3$ at $H > 200$ Oe, the $\Delta Q(H)$ plot (curve 2) can be attributed only to the changes of the inelastic scattering of the N₂ molecules, due to the influence of the field on the state of the surface coated with the monolayer of chemisorbed oxygen. From the analogous plots of $\Delta Q(-H)$ at $H_0 = -3.5$ kOe and $H_0 = 3.5$ kOe (curves 4 and 5) it follows that, within the limits of errors, $\Delta Q(-H, \pm H_0) = \Delta Q(+H, \mp H_0)$. When the pressure is in-

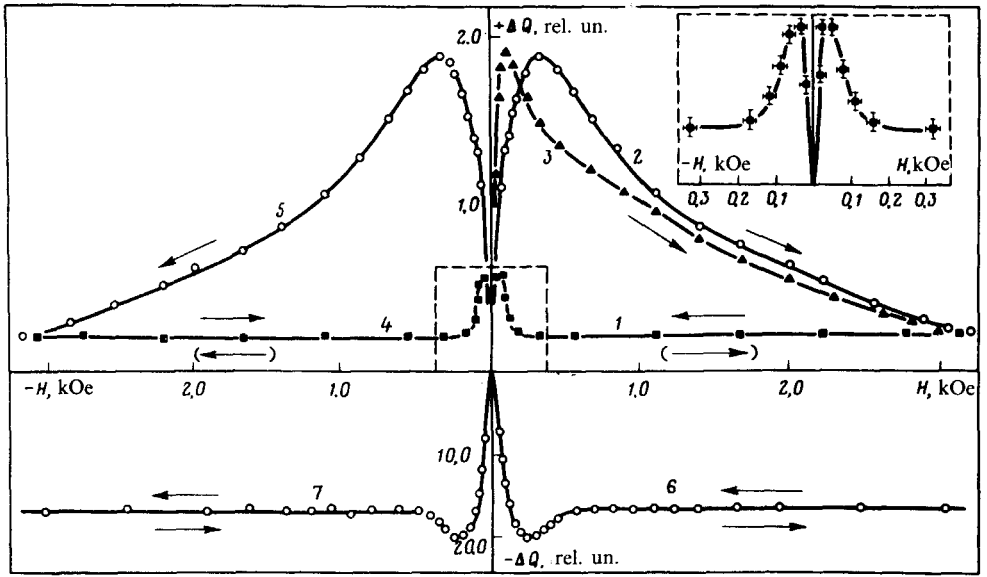


FIG. 1. Plots of the heat flux ΔQ following application of a magnetic field $\pm H$: $P=1 \times 10^{-3}$ Torr, curves 1-5— $T_c=77$ K, $T_g=130$ K, 6-7— $T_c=300$ K, $T_g=400$ K. Curve 1— $H_0=3.5$ kOe, 2— $H_0=-3.5$ kOe, 3— $H_0=0$, 4— $H_0=-3.5$ kOe, 5— $H_0=3.5$ kOe, 6,7— H_0 arbitrary.

creased to $P=10^{-1}$ Torr, when $\lambda \sim 0.1L$, the plots 1-5 agree with one another and with the plot typical of the Senftleben effect^[4]; this also attests to the influence of the field on the state of the surface.

The effect of the field on the state of the surface is confirmed by experiments in which the measured quantity δQ is connected only with the change in the state of the surface after a brief application of the field. The ordinates in Fig. 2 represent the difference $\delta Q=Q_1-Q_2$ between the fluxes after a brief application of the field $H_0=3.5$ kOe (Q_2) and after a subsequent brief application of the field $-H$ (the flux Q_1) [$\delta Q(H)$ remains unchanged when the signs of H_0 and H are simultaneously reversed]. The abscissas represent the field $-H$. It is seen that $\delta Q < 0$, goes through a minimum with increasing H (at $H \cong 500$ Oe), and then tends to zero. Since $\delta Q \approx 0$ at $H > 3.0$ kOe, the states of the surface at $H_0 = \pm 3.5$ kOe are shown by the molecule scattering to be identical. Therefore the plots of $\Delta Q(\pm H)$ (curves 1 and 4 on Fig. 1) can be regarded as corresponding to unchanged states of the surface at saturation. Curves 2 (5) can be connected with the change of Q when the magnetization is reversed from the state corresponding to $H_0 = -3.5$ kOe (3.5 kOe) into an identical state at $H_0 = 3.5$ kOe (-3.5 kOe). Since $\delta Q < 0$, a magnetically disordered state of the surface corresponds to a smaller heat flux.

The results shown in Fig. 1 (the direction of the evolution of the field H is indicated by the arrows) attests to hysteresis of the state of the surface.

The experiments have shown also that the result of the action of a field of definite magnitude and direction does not change after the pickup chamber is evacuated to

$P \leq 10^{-6}$ Torr and the surfaces are heated to $T \sim 350$ K. This removes the physically adsorbed molecules from the gold surface. Since the lifetime of the chemisorbed oxygen at this temperature on the gold surface is large, it can be assumed that the state measured in the field is precisely that of a monolayer of oxygen.

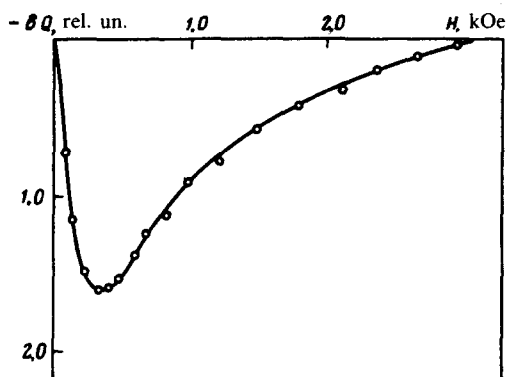


FIG. 2. Plot of $\delta Q(H)$: $T_c = 77$ K. $T_g = 130$ K.

It follows from the presented data that a change of the magnetic state of the surface leads to a change of the nonspherical scattering of only "cold" polarized molecules. This is confirmed by the fact that in argon ($P = 1 \times 10^{-3}$ Torr) we have $\delta Q = 0$, and also by the reversal of the sign of ΔQ and by the vanishing of the hysteresis with increasing temperature (see curves 6 and 7 on Fig. 2). As shown in^[2], $\Delta Q_{\text{sat}} > 0$ if the probability of nonspherical scattering changes with changing sign of the vector normal to the surface, and also of the vectors of the velocity and angular momentum of the incident and reflected molecules. This can take place if a preferred direction exists on the surface. We note that the above-described regularities of the influence of the field on the states of the monolayer of oxygen on the gold surface take place also in a field $\mathbf{H} \parallel \mathbf{k}$, but a definite anisotropy of the ferromagnetic properties is observed.

In conclusion, it is our pleasant duty to thank Yu. M. Kagan and L. A. Maksimov for a useful discussion and valuable remarks.

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