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ANTIFERROMAGNETIC PROPERTIES OF COBALTOUS OXIDE

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The magnetic susceptibility of CoO was investigated between 100 and 700°K. A sharp maximum was found at 292°K. The dependence of susceptibility on field in the antiferromagnetic state and in the region of the antiferromagnetic Curie point was determined.

1. One of the characteristic properties of antiferromagnets is the slow increase in magnetic susceptibility with increasing temperature, below the antiferromagnetic Curie point (Θ_{af}), and its decrease above this temperature, according to the Curie-Weiss law. Landau[1] was the first to explain theoretically this variation of magnetic susceptibility with temperature for antiferromagnets.

In the present work we have studied the temperature dependence of the magnetic susceptibility of polycrystalline specimens of cobaltous oxide. In other studies[2-4] of this substance it was shown that CoO has antiferromagnetic properties, but a sharp maximum in the susceptibility-temperature curve was only found in La Blanchetais' work.[4] Singer[5] studied the magnetic properties of a CoO single crystal and also found a susceptibility maximum. In all the work mentioned, the dependence of magnetic susceptibility χ on the strength of the magnetic field was hardly examined. The existing data on the dependence of χ on H for other antiferromagnets are inconsistent[4,6,7] and these facts have not yet been completely explained. It therefore seemed of great interest to study the magnetic susceptibility of cobaltous oxide over a wide range of magnetic field and for different temperatures.

2. The measurements of susceptibility were made with an apparatus based on the Faraday-Sucksmith method.[8] In this method, the force acting on a specimen, placed in a strong inhomogeneous magnetic field, is measured by the deformation of an elastic ring made of beryllium bronze. For a specimen mass of 10 — 25 mg the sensitivity of the apparatus was a susceptibility of $\sim 10^{-7}$ per scale division.

Cobaltous oxide was investigated over the temperature range 100 — 700°K. The low temperatures were obtained by using nitrogen vapor which flowed around the specimen under examination. A small nichrome heater was placed in a dewar containing liquid nitrogen, and its temperature determined the rate at which the nitrogen evaporated. The nitrogen vapor passed from the dewar vessel into a double-walled glass tube in which the specimen was hung freely on a quartz thread. A copper tube of smaller size was placed inside the glass tube so that there should be a uniform temperature over a certain length. Temperatures above room temperature were obtained by using a small oven wound of bifilar platinum wire on a porcelain tube of diameter 6 mm and length $\sim$ 70 mm. The temperature was measured with a copper-constantan thermocouple, previously calibrated.

The results of La Blanchetais[4] show that the magnetic properties of cobaltous oxide depend markedly on the method of preparing the specimens. For this reason great attention was paid

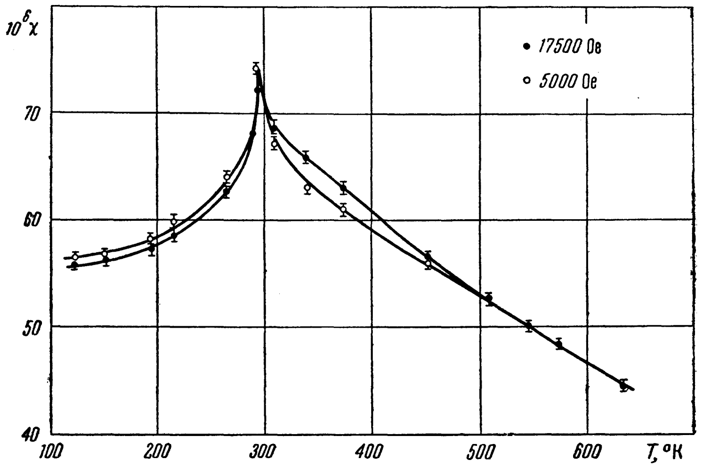

FIG. 1. The temperature dependence of magnetic susceptibility.

to specimen preparation. Cobalt nitrate was used, heated in air to 1000° C for three hours. Specimens were pressed from the powder so obtained under a pressure of 2000 kg/cm^2, and were heated in vacuum (10^{-3} mm Hg) for three hours at 820° C. We should remark that specimens prepared in this way were practically identical in magnetic properties.

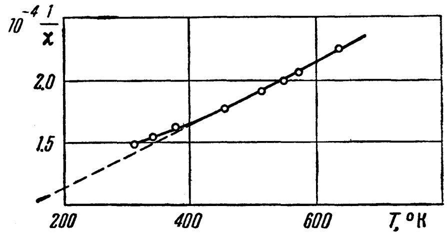

FIG. 2. The temperature dependence of the inverse magnetic susceptibility, H = 17,500 oe.

3. As stated above, the magnetic susceptibility was measured over the wide temperature range from 100 to 700°K. The temperature dependence of the specific susceptibility remains practically constant at low temperatures. With increasing temperature it increases steadily and has a sharp maximum at 292° K. With further increase of temperature χ rapidly decreases.

To determine the dependence of χ on H, the susceptibility was measured for various values of the magnetic field up to ~ 18,000 oe. Below the antiferromagnetic Curie point, i.e., in the antiferromagnetic state, the susceptibility decreases with increasing field, while at temperatures above Θ_{af}, near the transition point, χ on the contrary increases.

La Blanchetais[4] found the same dependence of χ on H for cobaltous oxide, but in her work the range of fields was small, so that the behavior was less marked. It is interesting that the difference between the susceptibilities for different fields decreases on approaching the Curie point and increases with decreasing temperature; according to our data this difference is greatest at T = 100° K.

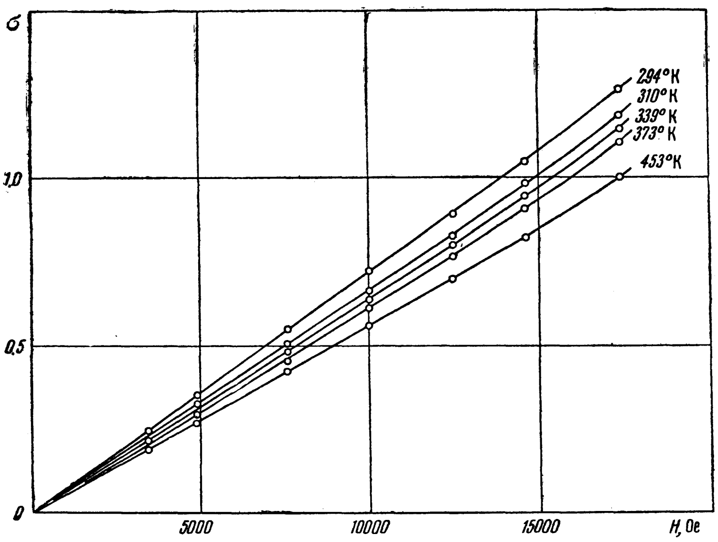

FIG. 3. Magnetization curves at temperatures above Θ_{af}.

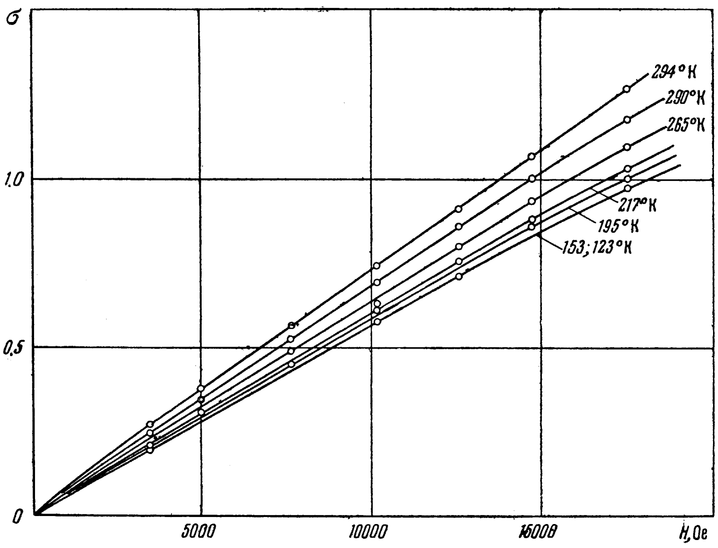

FIG. 4. Magnetization curves at temperatures below Θ_{af}.

We should point out that the temperature of the antiferromagnetic Curie point, $\Theta_{af} = 292°K$ is independent of the strength of the external field. The value of Θ_{af} obtained agrees well with the results of measurements of other physical quantities.[9]

The susceptibility at absolute zero can be obtained by extrapolation of the curve of the temperature dependence of susceptibility shown in Fig. 1 to $T = 0°K$. This is $\sim 55 \times 10^{-6}$ from our measurements. According to the theory of Van Vleck,[10] the susceptibility at $T = 0°K$ of an antiferromagnet with two sublattices is $2/3$ of the susceptibility at $T = \Theta_{af}$. For the case of several sublattices the value of χ_0/χ_{af} should be somewhat higher according to Anderson;[11] in fact we found $\chi_0/\chi_{af} = 0.76$.

Figure 2 shows the temperature dependence of $1/\chi$ above the antiferromagnetic Curie point. It can be seen that this dependence follows the Curie-Weiss law above 470°K. The values of the Curie-Weiss constant C, the atomic magnetic moment P_p and the paramagnetic Curie point Θ_p determined from our experimental data were $C = 3.05$, $P_p = 4.97$, $\Theta_p = -270°C$. The values of magnetic moment and of the paramagnetic Curie point agree well with the data of La Blanchetais[4] who obtained $P_p = 4.96$, $\Theta_p = -280°C$.

There is a temperature region above the antiferromagnetic Curie point ($T \gtrsim \Theta_{af}$) up to $\sim 470°K$ where the magnetic susceptibility is a function both of the temperature and of the magnetic field strength, as was also found by La Blanchetais. The isothermal magnetization curves in the antiferromagnetic state and in the region of the antiferromagnetic Curie point are given in Figs. 3 and 4. Near Θ_{af} the dependence of specific magnetization on H is practically linear. On lowering the temperature in the antiferromagnetic state, the isotherms start to curve gradually. The curves are then concave to the H axis. In the paramagnetic region ($T \gtrsim \Theta_{af}$) there is an analogous small curvature, but in the opposite direction.

4. The study of the temperature dependence of the magnetic susceptibility of cobaltous oxide showed that this substance takes on well defined antiferromagnetic properties, which appear at $T = 292°K$ in the form of a sharp maximum in the $\chi(T)$ dependence. In the paramagnetic region the susceptibility of CoO follows the Curie-Weiss law, which is observed in most antiferromagnets. A small dependence of magnetic susceptibility on field strength was found both above and below the temperature of the antiferromagnetic transition.

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