much as in Refs. 19 and 20 they investigated only the linear stage of the instability, and the thermal drift was not taken into account in the numerical experiment of Ref. 21, so that doubts are case on the large magnetic fields obtained in the latter reference.

The authors thank E. P. Velikhov, A. A. Samarskiĭ, and L. P. Feokistov for a discussion of the results.

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Translated by J. G. Adashko

Investigation of a magnetic phase transition in a layer of carbon monoxide molecules chemisorbed on platinum and gold surfaces in a magnetic field

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(Submitted 17 May 1979)
Zh. Eksp. Teor. Fiz. **77**, 2297–2312 (December 1979)

The thermomagnetic effect was used to study a magnetic phase transition in a monolayer of chemisorbed carbon monoxide molecules. A study was made of the dependence of the critical field H_c, in which the magnetic phase transition took place, on the surface concentration of the carbon monoxide molecules x_{CO} and the surface temperature T. A phase transition was also observed in a monolayer of chemisorbed hydrogen on the surface of platinum. An analysis was made of a change in the nonspherical interaction between the molecules and the surface which occurred at the phase transition. The observed dependences of H_c on x_{CO} and T could be interpreted as a consequence of a transition in a monolayer of chemisorbed particles from an antiferromagnetic to a ferromagnetic state in a field $H = H_c$.

PACS numbers: 75.30.Kz, 73.90. + f

1. INTRODUCTION

Numerous investigations of the adsorption on the surfaces of solids have established that, in addition to the interaction between the adsorbed atoms or molecules and the surface atoms of the crystal lattice of the adsorbent, there are also forces acting between the adsorbed atoms or molecules. The existence of a strong interaction between chemisorbed atoms or molecules is supported by, for example, the concentration (coverage) dependences of the heat of adsorption and electron work function.[1] Low-energy electron diffraction investigations have shown that this type of interaction can produce a monolayer with a geometrically ordered structure.[1] The collective interaction of adsorbed atoms and molecules can also result in magnetic ordering in the adsorption monolayer. As demonstrated in our earlier investigation,[2] a system of this kind may exhibit magnetic phase transitions induced by a magnetic field. A phase transition in a monolayer of chemisorbed carbon monoxide molecules on gold and platinum surfaces was observed by us earlier[2] with the aid of the thermomagnetic effect in a Knudsen molecular gas.

The thermomagnetic effect[3,4] is the change, in a magnetic field H, in the heat flow Q through a molecular gas located in a gap between two parallel surfaces $T_1 > T_2$ and characterized by a mean free path λ of molecules much greater than the gap L (Knudsen number $Kn = \lambda/L \gg 1$).

It is shown in Refs. 3 and 4 that the thermomagnetic effect is due to the nonspherical interaction of the gas molecules with the surface, which results in their polarization along the direction of the angular momentum **M**. In the presence of a magnetic field the molecular precession causes partial destruction of this polarization, resulting in averaging of the orientations **M** in a plane perpendicular to the field direction. This alters the distribution function of the molecules in the gap and, consequently, the heat flow. The parameter which determines the change in the heat flow ΔQ is the product $\omega\tau = \gamma HL/\bar{v}$ (ω is the precession frequency, τ is the transit time from one wall of the gap to the other, and γ is the gyromagnetic ratio of the molecules). On increase in $\omega\tau$ the value of ΔQ reaches its maximum (at $\omega\tau \sim 1$) and then tends to a limiting (saturation) value $\Delta Q^{\text{sat}} = \text{const}$.

The change in the energy flow through the gas due to the application of a magnetic field may be not only due to the precession of the molecules but also due to a change in the inelastic scattering of molecules on the surface because the field affects the state of the surface. In the collisionless regime of heat transfer the gas molecules in the gap act as probe particles whose polarization depends on the state of the surface.

The present paper is devoted to a magnetic phase transition discovered earlier[2] in a monolayer of carbon monoxide chemisorbed on gold and platinum surfaces. The experimental results are given in Sec 2. We studied the dependences of the critical field H_c in which the transition took place on the concentration of carbon monoxice on the surface. It was found that H_c depended on the surface temperature. A magnetic phase transition was observed also in a monolayer of hydrogen chemisorbed on the surface of platinum. The theory of the thermomagnetic effect was used to analyze how the magnitude of this effect was affected by a change in the orientational scattering of molecules by the surface resulting from the magnetic phase transition (Sec. 3).

2. EXPERIMENTAL RESULTS

Our experiments were carried out using apparatus similar to that employed earlier.[4] A heat flow was established between the "hot" surface and two "cold" surfaces located symmetrically, at a distance of 0.2 cm, on both sides of the hot surface. This hot surface was a mica plate 5×10^{-3} mm thick and of 40×50 mm dimensions which had a film of gold or platinum (with impurity concentrations ≤ 0.01 at.%) evaporated on both sides. The cold surfaces were polishd brass plates coated with the same type of evaporated gold or platinum film.

An analysis of an evaporated gold film, carried out using an x-ray photoelectron spectrometer (the film was probed to a depth of 30–40 Å), showed that the surface layer of the film did not contain (to within 1%) magnetic metal impurities. The photoelectron spectrum had not only peaks associated with various electron shells of gold but also oxygen and carbon peaks, indicating the presence of oxygen and carbon impurities on the surface. The crystal structure of the gold film was determined with an x-ray diffractometer. It was found that the grains in the evaporated film were oriented mainly so that their $\{111\}$ faces were parallel to the surface of the hot element.

Nonmagnetic materials were used in this system, which was placed inside a vacuum chamber and the chamber was placed inside an electromagnet gap. A special rotation mechanism made it possible to alter the orientation of the field and to carry out measurements in fields $\mathbf{H} \perp \mathbf{k}$ or $\mathbf{H} \parallel \mathbf{k}$ (**k** is the normal to the surface). The gold or platinum film on mica acted as a thermistor connected to a measurng circuit and it was heated by a dc current. Measurements were made of the change in the resistance of this film due to the change, in a magnetic field, of the energy flux in the gas because of the precession of the molecules and also because of the change in the inelastic scattering of the molecules by the surface when its state was modified by the magnetic field. The measurements were carried out at a gas pressure of $p = 1\times10^{-3}$ Torr, which corresponded to a Knudsen number $Kn \gtrsim 20$ (at $T = 350$°K.

The composition of a chemisorbed layer on a metal surface was altered by heterogeneous catalytic reactions of the oxidation of CO and H_2 by oxygen.[5,6] At temperatures $T \leq 500$°K, carbon monoxide was chemisorbed on the surface of platinum in the form of molecules, whereas oxygen and hydrogen were in the form of atoms.[6] Hydrogen was not chemisorbed on the gold surface.[6] Oxygen and carbon monoxide on gold were chemisorbed in the form of atoms and molecules, respectively.[6] A preliminary check was made of the catalytic activity of evaporated gold and platinum films. A stoichiometric mixture of $2H_2 + O_2$ (or $2CO + O_2$) at a pressure of $p = 1\times10^{-1}$ Torr was admitted to a probe chamber where both surfaces were heated to 400–550°K. The catalytic reaction (reaction products CO_2 and H_2O were frozen out in a liquid-nitrogen trap) was monitored by recording the fall of the pressure under steady-state conditions or by measurements with a PMT-2 manometric tube or by determining the thermal resistance of the probe. The catalytic activity of the gold surface was very low compared with the activity of the platinum surface and the maxinum saturation time of the surface with the carbon monoxide molecules was ≈60 h.

The reported measurements showed that the change in ΔQ associated with the replacement of certain particles with others (for example, with the replacement of carbon monoxide with oxygen) in a chemisorbed

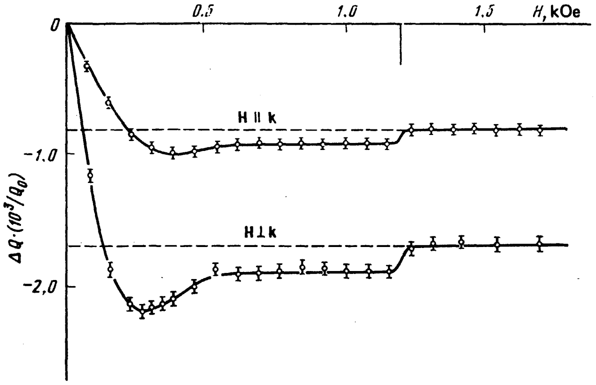

FIG. 1. Plots of $\Delta Q(H)$ for the CO/(Au—Au)+CO system ($p = 1\times10^{-3}$ Torr, T_c 298°K, $T_h = 383$°K).

monolayer were reversible.

Under the experimental conditions employed in our study ($p \sim 1\times10^{-3}$ Torr, $T_{av} \approx 350$°K), the physical sorption of gases on the metal surfaces could be ignored. Estimates obtained using the Brunauer-Emmett-Teller (BET) method[7] showed that in the case of physical-sorption energies of, for example, $E_a = 5$ kcal/mole for nitrogen[8] the surface coverage would be $\theta \sim 10^{-4} \ll 1$.

The thermomagnetic effect was investigated in nitrogen and carbon monoxide interacting with platinum and gold surfaces subjected, before the measurements, to a treatment in an atmosphere of oxygen, carbon monoxide, or hydrogen gases in an accordance with the above method. Figure 1 shows the dependences $\Delta Q(\mathbf{H})$ obtained for carbon monoxide interacting with gold surfaces saturated with chemisorbed carbon monoxide gas [CO/(Au–Au) + CO system]. It is clear from Fig. 1 that when the critical field was $H_c = 1200$ Oe, there was an abrupt change in the heat flow

$$\gamma = \Delta Q^{\mathrm{sat}}(H>H_c) - \Delta Q^{\mathrm{sat}}(H<H_c). \qquad (1)$$

In the case of different orientations of the H ⊥ k and H ∥ k fields, it was always found that $H_c^{\perp} \approx H_c^{\parallel}$. The magnitude of the jump γ was in both cases ~10% of ΔQ^{sat}.

At a fixed temperature of the hot surface T_h and for saturation of the gold surface with chemisorbed carbon monoxide molecules the value $H_c \approx 1150$ Oe was a characteristic of the system and did not change (within the limits of the experimental error) when the thermal elements were interchanged.

Our experiments indicated that the abrupt change in the heat flow was observed only in the Knudsen regime ($Kn \gg 1$), when the transfer of heat was due to collisions of the gas molecules with the surfaces. At higher gas pressures in the gap the abrupt change in the heat flow disappeared and the dependences $\Delta Q^{\perp}(H)$ and $\Delta Q^{\parallel}(H)$ became monotonic, typical of changes in the thermal conductivity in a magnetic field (Sentftleben effect[9]).

It follows from the theory of the thermomagnetic effect[4] that an increase in the parameter $\omega\tau$ causes the magnitude of the effect $\Delta Q(\omega\tau)$ to approach a constant value $\Delta Q^{\mathrm{sat}} = \mathrm{const}$. This value ΔQ^{sat} is governed by the nature of the nonspherical interaction of the molecules with the solid surface. Moreover, ΔQ^{sat} depends, for a fixed geometry of the experiment, on the difference between the wall temperatures, gas pressure, and field orientation. Hence, it follows that the discontinuity in the heat flow observed in a field $H = H_c$, when $\omega\tau \gg 1$, is related to the change in the nonspherical interaction of the gas molecules with the surface resulting from the field-induced change in the state of the chemisorbed monolayer of carbon monoxide. This conclusion was supported by the results of an experiment in which carbon monoxide was replaced with nitrogen. Once again an abrupt change in the heat flow was observed and the field $H_c \approx 1200$ Oe agreed, within the limits of the experimental error, with the value deduced from Fig. 1. Before these nitrogen experiments the gold surface was treated exactly in the same way an in the carbon monoxide experiments.

Since the earlier[2] investigation of the thermomagnetic effect in the N_2/(Au–Au) + O system failed to reveal a jump in the heat flow, it seemed of interest to study the dependences of γ and H_c on the carbon monoxide concentration in the adsorption layer. This concentration was altered by varying the duration of the preliminary treatment of the probe surfaces in the atmosphere of this gas.

The dependences $\Delta Q(\mathrm{H})$ obtained for different carbon monoxide concentrations on the gold surface are plotted in Fig. 2. We can see that in the case of surfaces saturated with the carbon monoxide molecules the jump occurs at $H_c = 1200$ Oe (curve 1). When carbon monoxide is partly replaced with oxygen (as a result of an additional treatment in an oxygen atmosphere), the value of H_c decreases (curve 2). When the gold surfaces are saturated with oxygen (curve 3), the dependence $\Delta Q(\mathbf{H})$ agrees with that given in Ref. 4. It should also be noted that a reduction in the proportion of the carbon monoxide molecules in the adsorption layer tends to reduce also the value of γ.

Similar dependences $\Delta Q(\mathrm{H})$ were obtained for various concentrations of carbon monoxide in the chemisorbed layer also when the probe gas was carbon monoxide [CO/(Au–Au) + CO system].

The dependence of H_c on the duration t of treatment of the gold surface in a carbon monoxide atmosphere is plotted in Fig. 3. We can see that the value of H_c reaches saturation after $t > 60$ h. The dependence of

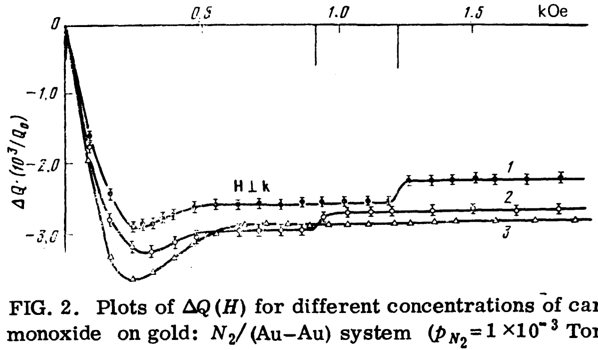

FIG. 2. Plots of $\Delta Q(H)$ for different concentrations of carbon monoxide on gold: N_2/(Au–Au) system ($p_{N_2} = 1\times10^{-3}$ Torr, $T_c = 298$°K, $T_h = 383$°K).

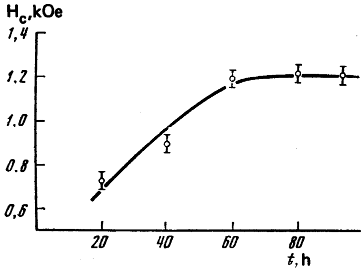

FIG. 3. Plots of $H_c(t)$ for the CO/(Au–Au) system ($p_{CO}=1\times10^{-3}$ Torr, $T_c=298$ °K, $T_h=383$ °K).

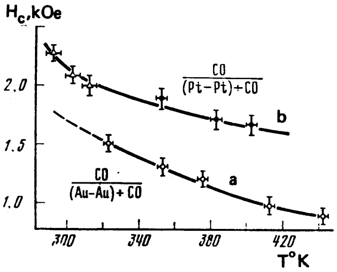

FIG. 5. Plots of $H_c(T)$: a: $T_c=298$ °K; b: △) $T_h=383$ °K, ●) $T_c=298$ °K.

H_c on the carbon monoxide concentration on the gold surface should be accompanied by an associated temperature dependence of H_c, because the monolayer coverage (carbon monoxide concentration) depends on the surface temperature.

Since the heat flow Q through a gas in a gap between two surfaces kept at different temperatures is governed by the processes of energy transfer at both surfaces, there should be two jumps in the heat flow corresponding to the field-induced changes in the chemisorbed layers of particles on the two surfaces. This was observed in a study of the thermomagnetic effect in the CO/(Au–Au) +CO system (Fig. 4). The heat flow jumps γ_1 and γ_2 were observed in fields $H_c^{(1)}=1200$ Oe and $H_c^{(2)}=1800$ Oe. The values of γ_1 and γ_2 amounted to 10% of ΔQ^{sat}.

The existence of a temperature dependence of the critical field H_c was confirmed by a study of the thermomagnetic effect at various temperatures of the hot surface (the temperature of the cold surface was kept constant). Figure 5a shows the dependence of $H_c^{(1)}$ on the hot-surface temperature T_h. Extrapolation of the curve obtained to $T=300$°K gave $H_c=1700$ Oe, which was close to $H_c^{(2)}=1800$ Oe. Hence, we concluded that the first jump in the heat flow in the CO/(Au–Au) +CO system was due to the magnetic-field-induced change in the state of the layer of chemisorbed carbon monoxide on the hot surface, whereas the second jump was due to the corresponding change in the state of these molecules on the cold surface.

The jumps in the heat flow were also observed on platinum surfaces. Figure 6 shows the dependences ΔQ(H) obtained for the CO/(Pt–Pt) + CO system. It is clear from this figure that in fields $H_c^{(1)}=1700$ Oe and $H_c^{(2)}=2200$ Oe there are two jumps of the heat flow. These jumps amount to 10% of ΔQ^{sat} and the values of $H_c^{(1)}$ and $H_c^{(2)}$ coincide (within the limits of the experimental error) for the H ⊥ k and H ∥ k configurations. It should also be noted that the directions of the jumps in this system ($\gamma_1<0$, $\gamma_2>0$) and in the CO/(Au–Au) + CO system (γ_1, $\gamma_2>0$) are different (the directions of the jumps, i.e., the signs of γ_1 and γ_2, are discussed below).

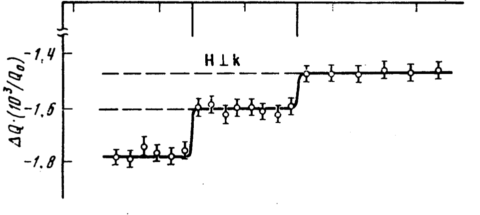

FIG. 4. Plots of $\Delta Q(H)$ for the CO/(Au–Au) + CO system ($p_{CO}=1\times10^{-3}$ Torr, $T_c=298$ °K, $T_h=383$ °K).

The temperature dependence of H_c for the CO/(Pt–Pt) + CO system is shown in Fig. 5b. The experiments were carried out as follows. A study was made of the dependence ΔQ(H) at a fixed temperature T_c of the cold surface but different value of T_h. There was a change in the value of $H_c^{(1)}$. Variation of T_c for a fixed T_h altered the value of $H_c^{(2)}$. From these results we concluded that the first jump was associated with the field-induced change in the chemisorbed layer of carbon monoxide on the hot surface, whereas the second jump was associated with the corresponding change on the cold surface.

Two jumps in the heat flow were also observed when the probe surfaces were made of different metals. Figure 7 shows the dependences ΔQ(H) obtained for carbon monoxide interacting with gold (hot surface) and platinum (cold surface), both saturated with chemisorbed carbon monoxide molecules [CO/(Au$_h$–Pt$_c$) + CO system]. It is clear from this figure that there are two jumps in the heat flux and that $\gamma_1>0$, whereas $\gamma_2<0$.

The values of γ_1 and γ_2 amount to 10% of ΔQ^{sat}. A comparison of the dependences ΔQ(H) in Figs. 4, 6, and 7 shows that the first jump in the CO/(Au$_h$–Pt$_c$) + CO system corresponds to a field-induced change in the state of the layer of chemisorbed carbon mon-

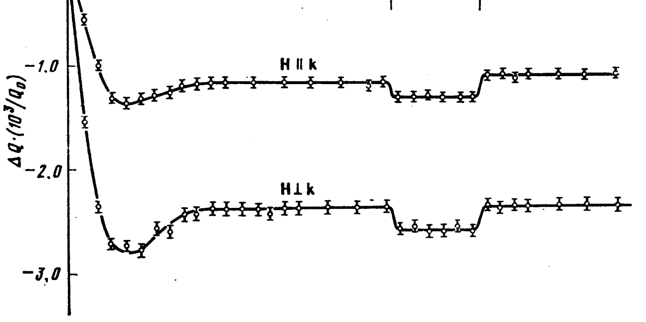

FIG. 6. Plots of $\Delta Q(H)$ for the CO/(Pt$_h$–Pt$_c$) + CO system ($p_{CO}=1\times10^{-3}$ Torr, $T_c=298$ °K, $T_h=383$ °K).

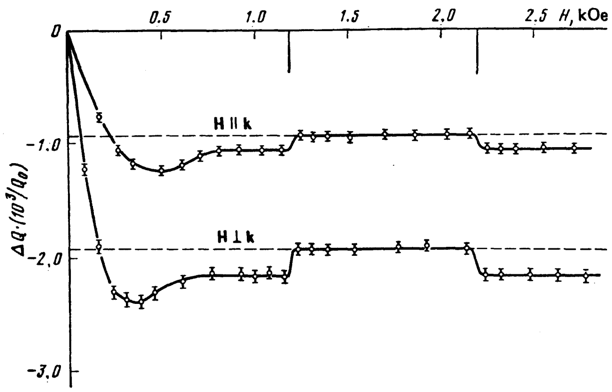

FIG. 7. Plots of $\Delta Q(H)$ for the CO/ ($Au_h - Pt_c$) + CO system ($p_{CO} = 1 \times 10^{-3}$ Torr, $T_c = 298$ °K, $T_h = 383$ °K).

oxide molecules on the gold surface and the second corresponds to a similar change on the platinum surface.

Adsorption on metal surfaces was also studied for other gases. It was found that a jump in the heat flux also occurred when chemisorbed hydrogen was present on platinum surfaces. Measurements carried out on the $N_2/(Au_h - Pt_c) + H_2$ system revealed only one jump (Fig. 8). An increase in the hydrogen concentration on the surface (i.e., an increase in the duration of the treatment of the surfaces in a hydrogen atmosphere) increased H_c from 2700 to 3500 Oe (Fig. 8). The subsequent treatment of the surfaces in a hydrogen atmosphere did not increase H_c. The absence of a dependence of H_c on the temperature T_c and the absence of chemisorption of hydrogen on gold (according to Trapnell[8]) led to the conclusion that the observed jump in the heat flow was due to a field-induced change in the state of a layer of chemisorbed hydrogen atoms on the platinum surface. As in the case of chemisorption of carbon monoxide, the jump γ amounted to 10% of ΔQ^{sat}.

3. DISCUSSION OF RESULTS

The dependence of H_c on the concentration of the carbon monoxide molecules on a solid surface and on the temperature of this surface, as well as the relatively small width of the jump ($\Delta H/H_c \leq 0.05$), indicating simultaneous changes in the states of molecules chemisorbed on different parts of the surface, indicated a collective magnetic interaction in the chemisorbed layer. Therefore, the transitions occurring on the surfaces of gold and platinum in a magnetic field can be regarded as phase transitions of magnetic origin and associated with a change in the spin structure of the carbon monoxide layer. The occurrence of a magnetic phase transition can be explained by postulating the antiferromagnetic interaction in a quasitwo-dimensional layer of carbon monoxide or hydrogen chemisorbed on gold or platinum. However, the abrupt change in the state of a chemisorbed monolayer can be explained only if we assume also an anisotropy of the antiferromagnetic interaction becaue isotropic antiferromagnetics do not exhibit sudden changes in the magnetic moment under the action of an external magnetic field.[10] The anisotropy in this case is clearly due to the structure of the surfaces of gold and platinum single crystals on the surfaces of the evaporated films of these metals.

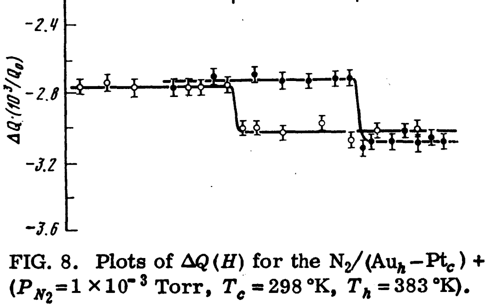

FIG. 8. Plots of $\Delta Q(H)$ for the $N_2/(Au_h - Pt_c) + H_2$ system ($P_{N_2} = 1 \times 10^{-3}$ Torr, $T_c = 298$ °K, $T_h = 383$ °K).

A special feature of the observed magnetic phase transition is its occurrence in weak fields $H_c \sim 10^3$ Oe and at relatively high temperatures $T \sim 300$°K. The fact that the sudden change in the magnetic moment, associated with the destruction of the antiferromagnetic order by the applied field, can occur in $H \ll kT/\mu_{eff}$ (T is the surface temperature and μ_{eff} is the effective magnetic moment of an atom) was explained many years ago by Néel[11] in the case of layer antiferromagnets. It follows from Néel's analysis that, for certain ratios of the anisotropy constants and the exchange antiferromagnetic interaction energy, a magnetic system is in an antiferromagnetic state which experiences a sudden change in the magnetic moment in an external magnetic field parallel to the antiferromagnetic axis and goes over to a ferromagnetic state.

It has been established experimentally that H_c increases on increase in the concentration of the carbon monoxide molecules on the surface of gold or platinum up to a certain maximum value H_c^0 (Fig. 3), which clearly corresponds to the maximum possible, at a given temperature, coverage of a monolayer with chemisorbed carbon monoxide molecules. The experimentally determined dependences of this field on the temperatures of the gold or platinum surfaces (Fig. 5) also indicate that the antiferromagnetic exchange interaction decreases on increase in the distance between the chemisorbed carbon monoxide molecules. The case of a strong dependence of the antiferromagnetic exchange interaction on this distance was considered by Kittel.[12] Kittel's model of the exchange inversion is based on the assumption that the exchange interaction between two sublattices with the magnetizations $\mathbf{M}_A$ and $\mathbf{M}_B$ is a linear function of the lattice parameter and it changes sign at a certain value of this parameter. A comparison of the experimental temperature dependence of the field with the corresponding dependence obtained by Kittel in Ref. 12 makes it possible to interpret the observed change in the state of an absorption layer as a field-induced transition from a antiferromagnetic to a ferromagnetic state. It follows that an increase in the concentration of carbon monoxide at fixed values of H and T should result in a transition from a ferromagnetic to an antiferromagnetic state.

We shall now analyze the relationships between the jumps in the heat flow and their signs observed in different systems. It should be noted that the ratios of the magnitudes of the jumps to the value of ΔQ^{sat} can be regarded as the same (within the limits of the experimental error) for the $\mathbf{H} \parallel \mathbf{k}$ and $\mathbf{H} \perp \mathbf{k}$ configurations. This shows that it is the nonsphercial part of the interaction of the molecules with the surface that changes. Such an analysis allows us to determine the properties of the nonspherical scattering probability which change as a result of change in the state of the surface in a field H_c.

The expression for the dependence of the change in the heat flow between two plane-parallel surfaces (one hot and one cold) on the magnetic field in a Knudsen polyatomic gas is obtained in Ref. 4. The effect is described by solving the transport equation with boundary conditions on the surfaces. These conditions relate the distribution functions of the incident f^- and reflected f^+ (by the surface) molecules, and are expressed in the form of an integral relationship whose kernel is the density of the probability of the surface scattering of the molecules $W(\mathbf{v}'\mathbf{M}' \to \mathbf{v}, \mathbf{M}; \mathbf{k})$. This scattering probability can be represented in the form

$$W = W_0 + \varepsilon W_1, \tag{2}$$

where W_1 is the nonspherical part of the probability W and ε is a small nonspherical parameter. The probability W_0 describes elastic specular and diffuse reflection, which is isotropic in respect of the directions of the vectors $\mathbf{v}$ and $\mathbf{M}$ of the emerging molecules.

The expression for the heat flow in a magnetic field can be obtained for an arbitrary nonspeherical interaction of molecules with a surface if we use an expansion of the function W_1 in terms of spherical harmonics of the vectors $\mathbf{M}'$ and $\mathbf{M}$:

$$W_1(\mathbf{v}', \mathbf{M}' \to \mathbf{v}, \mathbf{M}; \mathbf{k}) = \sum_{lnl'n'} \omega_{lnl'n'}(\mathbf{v}', \mathbf{M}'^2, \mathbf{v}, M^2; \mathbf{k}) Y_{l'n'}\left(\frac{\mathbf{M}'}{M'}\right) Y_{ln}\left(\frac{\mathbf{M}}{M}\right). \tag{3}$$

It is found[4] that in the expression for the heat flow there are only functions of the type $\omega_{ln00}(\mathbf{v}', M'^2, \mathbf{v}, M; \mathbf{k})$. In an analysis of a jump in the heat flow we shall use an expansion of this function (the direction of the z axis coincides with the normal to the lower plane) in the form

$$\omega_{ln00}(\Gamma', \Gamma, \mathbf{k}) = |\mathbf{v}\mathbf{k}| f_M(E) \sum_{L_1 l_3 l_1 l_1'} \alpha^{L_1 l l_3}_{l_1' l_1 l 0}(v'^2, v^2, M'^2, M^2) \cdot \left(\frac{M'}{M_0}\right)^l \begin{pmatrix} L_1 & l & l_3 \\ -n & n & 0 \end{pmatrix} Y_{l_3 0}(\mathbf{k}) \sum_{m_1 m_1'} \langle l_1' m_1' l_1 m_1 | L_1 - n\rangle Y_{l_1' m_1'}(\mathbf{v}') Y_{l_1 m_1}(\mathbf{v}). \tag{4}$$

The solution of the transport equation with the boundary conditions on each of the surfaces makes it possible to obtain, in the seoond order in ε, an expression for that part of the heat flow which depends on the magnetic field $Q_2(\mathbf{H})$:

$$\left.\begin{aligned} &Q_2(\mathbf{H}) = \frac{4\pi\varepsilon^2}{(2-\alpha)^2} \frac{\Delta T}{T_0^2} \iiint_{v_z, v_z', v_z'' > 0} \zeta' f_0(E') \\ &\times EE'' \sum_{lmn} \frac{(-1)^n}{\Delta_m'} [d_{mn}^l(\theta_H)] R_{lmn}(\omega^+, \omega^-) d\Gamma d\Gamma' d\Gamma'', \\ &\Gamma = (\mathbf{v}, M^2), \quad \zeta' = |v_z'|, \quad \Delta_m' = e^{imn\eta'} - (1-\alpha)^2 e^{imn\eta'}, \\ &\eta' = \gamma HL/\zeta'. \end{aligned}\right\} \tag{5}$$

Here, α is the diffuse reflection coefficient; $f_0(E)$ is the Maxwellian distribution with a temperature T_0; the functions $d^l_{mn}(\theta_H)$ (rotational transformation coefficients) are defined in Ref. 13; θ_H is the polar angle of the field orientation. The expression for $R_{lmn}(\omega^+, \omega^-)$ is of the form

$$\begin{aligned} R_{lmn}(\omega^+, \omega^-) &= \omega^+_{l-n}(\Gamma_3' \Gamma) \omega_{ln}^-(-\Gamma_3', -\Gamma'') \\ &+ \omega^-_{l-n}(\Gamma', \Gamma_3) \omega_{ln}^+(-\Gamma', -\Gamma_3'') \\ &- (1-\alpha) e^{imn\eta'} \omega^+_{l-n}(\Gamma_3', \Gamma) \omega_{ln}^+(-\Gamma', -\Gamma_3'') \\ &- (1-\alpha) e^{imn\eta'} \omega^-_{l-n}(\Gamma', \Gamma_3) \omega_{ln}^-(-\Gamma_3', -\Gamma''), \end{aligned} \tag{6}$$

where ω^+ applies to the hot surface and ω^- to the cold surface,

$$\Gamma_3 = (\mathbf{v}_3, M^2), \quad \mathbf{v}_3 = \mathbf{v} - 2\mathbf{k}(\mathbf{v}\mathbf{k}).$$

In sufficiently high fields ($\omega\tau \gg 1$) the quantitiy Q_2 ceases to depend on the field and assumes the value Q_2^{sat}. Splitting $\omega_{ln}(\Gamma', \Gamma; \mathbf{k})$ into parts which are symmetric and antisymmetric in respect of reflection in the horizontal plane, σ_n (i.e., $v_z' \to -v_z', v_z \to -v_z, \mathbf{k} \to -\mathbf{k}$), and with respect to rotation by 180° about the z axis, C_{2z} (i.e., $v \to -v_3 v' \to -v_3', \mathbf{k} \to \mathbf{k}$), we can write the heat flow Q_2^{sat} in the form

$$\begin{aligned} Q_2^{sat} = \frac{8\pi\varepsilon^2}{(2-\alpha)^2} \frac{\Delta T}{T_0^2} \iiint_{v_z, v_z', v_z'' > 0} \zeta' f_0(E') EE'' \\ \times \sum_{ln} (-1)^n [d_{0n}^l(\theta_H)]^2 \Big\{ [C_{l-n}(\Gamma_3', \Gamma) C_{ln}(\Gamma_3', \Gamma'') \\ - D_{l-n}(\Gamma_3', \Gamma) D_{ln}(\Gamma_3', \Gamma'')] \frac{1}{2-\alpha} + [K_{l-n}(\Gamma_3', \Gamma) K_{ln}(\Gamma_3', \Gamma'') \\ - F_{l-n}(\Gamma_3', \Gamma) F_{ln}(\Gamma_3', \Gamma'')] \frac{1}{\alpha} \Big\} d\Gamma d\Gamma' d\Gamma''. \end{aligned} \tag{7}$$

Bearing in mind the known symmetry properties of the spherical functions, we can easily establish that C_{ln}, D_{ln}, F_{ln}, and K_{ln} can also be written in the form of Eq. (4) where the indices l_1, l_1', l_3, and n assume only the values given in Table I in our earlier paper.[4]

A change in the state of a layer of chemisorbed particles on a hot surface in a field $H \geqslant H_c^{(1)}$ alters the nonspherical part of the probability of the scattering of molecules by this surface. This gives rise to the first jump of $\Delta Q(\mathbf{H})$. In the case of an analogous change on a cold surface ($H \geqslant H_c^{(2)}$) there is a second jump of $\Delta Q(\mathbf{H})$ as a result of the first jump $\delta_1 Q_2$ and as a result of both jumps $\delta_2 Q_2$ can be described by

$$\delta_1 Q_2 = Q_2^{sat}(H > H_c^{(1)}, \omega^+ + \delta\omega^+, \omega^-) - Q_2^{sat}(H < H_c^{(1)}, \omega^+, \omega^-), \tag{8}$$

$$\delta_2 Q_2 = Q_2^{sat}(H > H_c^{(2)}, \omega^+ + \delta\omega^+, \omega^- + \delta\omega^-) - Q_2^{sat}(H < H_c^{(1)}, \omega^+, \omega^-). \tag{9}$$

The quantities $\delta_1 Q_2$ and $\delta_2 Q_2$ are related to the above jumps in the heat flux γ_1 and γ_2 by the following expressions:

$$\left.\begin{array}{c}\delta_1 Q_2=\gamma_1,\quad \delta_2 Q_2=\gamma_1+\gamma_2,\\ \gamma_1=\Delta Q^{\mathrm{sat}}(H_c^{(1)}<H<H_c^{(2)})-\Delta Q^{\mathrm{sat}}(H<H_c^{(1)}),\\ \gamma_2=\Delta Q^{\mathrm{sat}}(H>H_c^{(2)})-\Delta Q^{\mathrm{sat}}(H_c^{(1)}<H<H_c^{(2)}).\end{array}\right\} \tag{10}$$

It has been found experimentally that in the case of the gold surfaces we have $\delta_1 Q_2 < 0$, $\delta_2 Q_2 > 0$, and $\delta_2 Q_2 \approx 2\delta_1 Q_2$. In the case of the platinum surfaces, we obtain $\delta_1 Q_2 < 0$ and $\delta_2 Q_2 \gtrsim 0$.

It is known from the experimental results that a sudden change in $\Delta Q(H)$ occurs in fairly high fields ($\omega\tau \gg 1$). It is clear from Eqs. (5)–(7) that for these fields the value of ΔQ^{sat} is governed only by the terms with $m = 0$.

It is convenient to begin an analysis of Eqs. (8) and (9) from the H ∥ k case. We then have $d^l_{0n}(\theta = 0) = \delta_{0,n}$. Consequently, if we apply Eq. (7) to the H ∥ k configuration, then Eqs. (8) and (9) contain only the functions $\omega_{l0}(\Gamma'_3, \Gamma)$, i.e., only the terms of the C_{l0} and F_{l0} type (see Table I in Ref. 4). Using Eq. (4) and the properties of the $3j$ symbols, we can show that $C_{l0} \sim \delta_{l,2k}$ and $F_{l0} \sim \delta_{l,2k+1}$. This makes it possible to separate contributions of terms of the C_{l0} and F_{l0} type in Eqs. (8) and (9).

Bearing this in mind, we can now rewrite the expressions for $\delta_1 Q_2$ and $\delta_2 Q_2$ in the form

$$\begin{gathered}\delta_1 Q_2=\hat{A}\{2\alpha C_{l0}(x)\delta C_{l0}(x'')-(1-\alpha)\delta C_{l0}(x)\delta C_{l0}(x'')\\ -2(2-\alpha)F_{l0}(x)\delta F_{l0}(x'')-(1-\alpha)\delta F_{l0}(x)\delta F_{l0}(x'')\}=\gamma_1,\end{gathered} \tag{11}$$

$$\begin{gathered}\delta_2 Q_2=\hat{A}\{4\alpha C_{l0}(x)\delta C_{l0}(x'')+2\alpha\delta C_{l0}(x)\delta C_{l0}(x'')\\ -4(2-\alpha)F_{l0}(x)\delta F_{l0}(x'')-2(2-\alpha)\delta F_{l0}(x)\delta F_{l0}(x'')\}=\gamma_1+\gamma_2,\\ x=(\Gamma_3'\Gamma),\quad x''=(\Gamma_3'\Gamma'').\end{gathered} \tag{12}$$

Here, the integral operator $\hat{A}$ is defined by

$$\begin{gathered}\hat{A}f_{ln}(\Gamma,\Gamma',\Gamma'')=\frac{4\pi e^2}{(2-\alpha)^2}\frac{\Delta T}{T_0^2}\iiint\limits_{v_z,v_z',v_z''>0}\zeta' f_0(E')\\ \times EE''\sum_{ln}\frac{(-1)^n}{\alpha(2-\alpha)}[d_{0n}{}^l(\theta_H)]^2 f_{ln}(\Gamma,\Gamma',\Gamma'')\,d\Gamma\,d\Gamma'\,d\Gamma''.\end{gathered} \tag{13}$$

Using the symmetry properties of the functions $C_{ln}(x)$ and $F_{ln}(x)$, we obtain the following relationships:

$$\hat{A}\delta C_{l0}(x)\delta C_{l0}(x'')\geqslant 0,\quad \hat{A}\delta F_{l0}(x)\delta F_{l0}(x'')\leqslant 0. \tag{14}$$

Multiplying Eq. (11) by 2 and subtracting the result from Eq. (12), and then applying Eq. (10), we obtain

$$\hat{A}\{2\delta C_{l0}(x)\delta C_{l0}(x'')-2\delta F_{l0}(x)\delta F_{l0}(x'')\}=\gamma_2-\gamma_1. \tag{15}$$

It is clear from Eqs. (14) and (15) that $\gamma_2 - \gamma_1 \geqslant 0$. The experimental accuracy is insufficient to determine the different values of the first and second jumps in the case of gold surfaces. However, if we assume that $\gamma_1 = \gamma_2$, then each of the terms on the left-hand side of Eq. (15) must be assumed to be zero. The consequence is a general absence of the jumps of $\Delta Q(H)$. Thus, we reached the conclusion that $\gamma_2 > \gamma_1$ should be satisfied for identical surfaces in the second approximation with respect to ε. Hence, we can conlude that if $\gamma_1 > 0$ then also $\gamma_2 > 0$. However, if $\gamma_2 < 0$, then $\gamma_1 < 0$ and $|\gamma_2| < |\gamma_1|$; when $\gamma_2 > 0$, γ_1 can have any sign if the condition $\gamma_2 > \gamma_1$ is obeyed. Hence, in the case of identical surfaces we cannot encounter a situation in which the first jump increases $\Delta Q(H)$ and the second reduces it, i.e., we should not have $\gamma_1 > 0$ and $\gamma_2 < 0$. In this case the inequality $\gamma_2 > \gamma_1$ is known to be disobeyed. The case when $\gamma_1 > 0$ and $\gamma_2 < 0$ can be realized only for different surfaces.

Using Eqs. (14) and (15) and bearing in mind that $\alpha \approx 0.8$, we easily obtain the following inequalitites

$$\left.\begin{array}{c}-0.1(\gamma_2-\gamma_1)\leqslant\hat{A}\{-(1-\alpha)\delta C_{l0}(x)\delta C_{l0}(x'')\\ -(1-\alpha)\delta F_{l0}(x)\delta F_{l0}(x'')\}\leqslant 0{,}1(\gamma_2-\gamma_1),\\ 0{,}8(\gamma_2-\gamma_1)\leqslant\hat{A}\{2\alpha C_{l0}(x)\delta C_{l0}(x'')\\ -2(2-\alpha)F_{l0}(x)\delta F_{l0}(x'')\}\leqslant 1{,}2(\gamma_2-\gamma_1).\end{array}\right\} \tag{16}$$

Hence, it follows that in the case of gold surfaces and H ∥ k, the contribution of the terms which are quadratic in respect of δC_{l0} and δF_{l0} is small so that if we ignore this contribution, Eqs. (11) and (12) can be rewritten in the form

$$\delta_1 Q_2=\hat{A}\{2\alpha C_{l0}(x)\delta C_{l0}(x'')-2(2-\alpha)F_{l0}(x)\delta F_{l0}(x'')\}, \tag{17}$$

$$\delta_2 Q_2=\hat{A}\{4\alpha C_{l0}(x)\delta C_{l0}(x'')-4(2-\alpha)F_{l0}(x)\delta F_{l0}(x'')\}. \tag{18}$$

The relationships (17) and (18) show that the jumps of $\Delta Q(H)$ should have the same direction and values, i.e., the relationship $2\delta_1 Q_2 = \delta_2 Q_2$ should be obyed. This is the relationship obtained in the experiments on gold surfaces ($\gamma_1 \approx \gamma_2$).

If we confine our attention to the smallest values of the index l in Eq. (7), then Eqs. (17) and (18) become

$$\delta_1 Q_2=\hat{A}\{2\alpha C_{20}(x)\delta C_{20}(x'')-2(2-\alpha)F_{10}(x)\delta F_{10}(x'')\}=\gamma, \tag{19}$$

$$\delta_2 Q_2=\hat{A}\{4\alpha C_{20}(x)\delta C_{20}(x'')-4(2-\alpha)F_{10}(x)\delta F_{10}(x'')\}=2\gamma. \tag{20}$$

In the case of the field confirguarion H ⊥ k, Eqs. (8) and (9) can contain only the functions ω_{ln} with even values of $(l - n)$ [since $d^l_{0n}(\pi/2) \sim \delta_{l-n,2k}$]. Therefore, in a field H ⊥ k, the terms F_{l0} make no contribution to the expressions for $\delta_1 Q_2$ and $\delta_2 Q_2$ [since $F_{l0} \sim \delta_{l,2k+1}$ and $d^l_{00}(\pi/2) \sim \delta_{l,2k}$]. We can show that if H ⊥ k, the jumps in ΔQ cannot be described if in addition to C_{20} we allow only for C_{40}. Hence, it follows that apart from terms of the C_{l0} and/or F_{l0} type, a change in the state of chemisorbed particles alters also other terms in the expansion of the nonspherical scattering probability with even values of $(l - n)$. If we confine ourselves to indices $l \leqslant 2$, then the terms in question are $C_{2\pm2}$, $F_{2\pm2}$, $K_{1\pm1}$, and $D_{1\pm1}$. The terms $C_{2\pm2}$ and $F_{2\pm2}$ do not give the corrrect form of the dependence ΔQ on H in the $H < H_c^{(1)}$ case and, therefore, they can be omitted in discussing a heat flow jump. On the other hand, allowance for the terms $D_{1\pm1}$ and/or $K_{1\pm1}$ makes it possible to describe the observed heat flow jumps in the H ⊥ k case.

We shall now consider the platinum surfaces. The experimental results show that

$$\delta_1 Q_2=-\gamma,\quad \delta_2 Q_2=s,\quad \gamma>0,\quad s\approx 0.2\gamma. \tag{21}$$

Using Eq. (21), we find that the expressions for $\delta_1 Q_2$ and $\delta_2 Q_2$ become

$$\begin{gathered}\delta_1 Q_2=\hat{A}_1\{2\alpha C_{l0}(x)\delta C_{l0}(x'')-(1-\alpha)\delta C_{l0}(x)\delta C_{l0}(x'')\\ -2(2-\alpha)F_{l0}(x)\delta F_{l0}(x'')-(1-\alpha)\delta F_{l0}(x)\delta F_{l0}(x'')\}=-\gamma,\end{gathered} \tag{22}$$

$$\begin{gathered}\delta_2 Q_2=\hat{A}\{4\alpha C_{l0}(x)\delta C_{l0}(x'')+2\alpha\delta C_{l0}(x)\delta C_{l0}(x'')\\ -4(2-\alpha)F_{l0}(x)\delta F_{l0}(x'')-2(2-\alpha)\delta F_{l0}(x)\delta F_{l0}(x'')\}=s.\end{gathered} \tag{23}$$

The system (22)–(23) yields

$$\hat{A}\{\delta C_{l0}(x)\delta C_{l0}(x'')-\delta F_{l0}(x)\delta F_{l0}(x'')\}=\gamma+s/2. \tag{24}$$

Hence, bearing in mind that $\alpha \approx 0.8$ and $s \approx 0.2\gamma$, we easily obtain the following inequalities

$$\left.\begin{array}{c}-0.22\gamma\leqslant\hat{A}\{-(1-\alpha)\delta C_{l0}(x)\delta C_{l0}(x'')-(1-\alpha)\delta F_{l0}(x)\delta F_{l0}(x'')\}\leqslant 0.22\gamma,\\ 1.78\gamma\leqslant\hat{A}\{2\alpha\delta C_{l0}(x)\delta C_{l0}(x'')-2(2-\alpha)\delta F_{l0}(x)\delta F_{l0}(x'')\}\leqslant 2.64\gamma,\\ -1.22\gamma\leqslant\hat{A}\{2\alpha C_{l0}(x)\delta C_{l0}(x'')-2(2-\alpha)F_{l0}(x)\delta F_{l0}(x'')\}\leqslant -0.78\gamma.\end{array}\right\}\tag{25}$$

The relationships (25) allow us to estimate the contribution of the terms which are linear and quadratic in respect of δC_{l0} and δF_{l0} made to the first and second jumps. It is clear from Eq. (25) that the discontinuous change in ΔQ(H) on the platinum surfaces subjected to a field H ∥ k can be described if the terms linear in δC_{l0} and δF_{l0} make negative contributions to $\delta_1 Q_2$ and $\delta_2 Q_2$, whereas the terms quadratic in δC_{l0} and δF_{l0} make a contribution approximately twice as large as the absolute value of the first jump of ΔQ(H).

If Eqs. (22) and (23) are simplifid by dropping the terms which are quadratic in δC_{l0} and δF_{l0}, the result is $\delta_2 Q_2 = 2\delta_1 Q_2$. This is in conflict with the experimental results on the platinum surfaces. If we ignore the terms linear in δC_{l0} and δF_{l0}, the solution of the system (22)–(23) does not satisfy the inequalities (14). If we confine ourselves to the indices $l \leqslant 2$, we find that the terms C_{20} and/or F_{10} change in fields $H > H_c^{(1)}$ and $H > H_c^{(2)}$ so that the relationships (22)–(25) are satisfied and this gives rise to the jumps ΔQ(H) observed for the platinum surfaces. The probabilities of the nonspherical scattering of the N_2 and CO molecules on the platinum surfaces found in Ref. 4 contain only these terms of the expansion (3).

In the H ⊥ k configuration the terms F_{10} are absent from the expressions for $\delta_1 Q_2$ and $\delta_2 Q_2$ $[d^1_{00}(\theta) = \cos\theta]$ and the contribution of the terms C_{20} decreases by a factor of 4. This gain indicates the presence of the term $\omega_{1\pm 1}$ in the expression for W_1 and its change in fields $H > H_c$.

The contribution of the terms quadratic in δC_{l0} and δF_{l0} to the expression for the jumps in the heat flow in the case of gold is small, which is in contrast to the case of platinum. It follows that there is only a small change in the probability of the nonspherical scattering of molecules on the surface of gold compared with the surface of platinum. Hence, we can conclude that in the case of the platinum surface in fields $H \geqslant H_c$ there is a greater change in the surface phonon spectrum. Moreover, it follows from Eq. (20) and the last of the inequalities in Eq. (25) that in the case of gold a change in the surface state increases the contribution of the terms C_{10} and/or H_{10} to W_1, whereas in the case of platinum there is a reduction in this contribution.

We shall now consider the experimental results obtained for a pair of different surfaces: gold (hot) and platinum (cold). It follows from the experiments that the first jump of ΔQ(H) is due to a change in the state of the hot surface of gold and increases ΔQ(H), whereas the second jump of ΔQ(H) is due to a change in the state of the cold platinum surface and it reduces ΔQ(H).

It is clear from the above analysis that this is the case which cannot occur in a system with the same surfaces. The nonspherical probability of the scattering of molecules on the gold and platinum surfaces differs only in respect of the weights of the individual terms in the expansion (3). Therefore, the expression for $\delta_2 Q_2$ in the H ∥ k configuration can be represented by

$$\delta_2 Q_2=\delta_1 Q_2{}^{\mathrm{Pt}}+\delta_1 Q_2{}^{\mathrm{Au}}+\hat{A}\{2\delta C_{l0}^{(1)}(x)\delta C_{l0}^{(2)}(x'')-2\delta F_{l0}^{(1)}(x)\delta F_{l0}^{(2)}(x'')\}.\tag{26}$$

The upper indices (1) and (2) refer to the gold and platinum surfaces, respeftively. The values of $\delta_1 Q_2^{\mathrm{Au}}$ and $\delta_1 Q_2^{\mathrm{Pt}}$ are governed by Eqs. (17) and (22). An analysis of the jumps of ΔQ(H) in the case of the gold surfaces shows that the last two terms in Eqs. (26) can be ignored. We then find that $\delta_2 Q_2$ is described by

$$\delta_2 Q_2=\delta_1 Q_2{}^{\mathrm{Au}}+\delta_1 Q_2{}^{\mathrm{Pt}}.\tag{27}$$

Moreoever, the expression for $\delta_1 Q_2$ is identical with Eq. (17) and, therefore, we can write down

$$\delta_1 Q_2=\delta_1 Q_2{}^{\mathrm{Au}}.\tag{28}$$

It is clear from Eqs. (27) and (28) that the first jump of ΔQ(H) in the (Au_h–Pt_c) system is analogous to the first jump in (Au_h–Au_c), whereas the second jump in (Au_h–Pt_c) is analogous to the first jump in (Pt_h–Pt_c). Thus, we may conclude that the abrupt change in the behavior of ΔQ(H) in the (Au_h–Pt_c) system can be described if we use the results obtained for (Au_h–Au_c) and (Pt_h–Pt_c). Moreover, these results allow us to predict the magnitude and direction of the jumps of ΔQ(H) in the (Pt_h–Au_c) system. We then find that:

$$\delta_1 Q_2=\delta_1 Q_2{}^{\mathrm{Pt}},\quad \delta_2 Q_2=\delta_1 Q_2{}^{\mathrm{Pt}}+\delta_1 Q_2{}^{\mathrm{Au}},\tag{29}$$

i.e., the first jump due to the change in $H > H_c^{(1)}$ of the state of the hot surface of platinum reduces ΔQ(H) and is analogous to te first jump in the (Pt_h–Pt_c) system whereas the second jump of ΔQ(H) due to a chnge in the state of the cold surface of gold increases ΔQ(H) and is analogous to the first jump in the system (Au_h–Au_c),

The mechanism of the influence of a magnetic phase transition in a chemisorbed molecular layer on the change in the heat flow is not yet fully understood. However, we can assume that this phase transition in an adsorbed layer alters the nature of the interaction both between the adsorbed molecules themselves and aslo between them and the substrate. This alters the spectrum of the surface phonons in the solid covered by a layer of carbon monoxide molecules. The corresponding changes of the inversion-invariant (C_{ln} and K_{ln}) and noninvariant (D_{ln} and F_{ln}) parts of the nonspherical probability of the scattering of molecules on the surface may result in an abrupt change in ΔQ(H) in the effect under consideration. The dependences $H_c^{(1)}(T_h)$ and $H_c^{(2)}(T_c)$ for the same surfaces agree within the limits of the experimental error. Hence, it follows that the value of the critical field in which a magnetic transition takes place in a layer of chemisorbed carbon monoxide molecules is determined entirely by the temperature of the surface (i.e., by the concentration of these molecules on the solid surface) and not by the average temperature during an experiment. Therefore, the gas molecules can be regarded as probe particles which do not affect the magnetic state of a layer of chemisorbed carbon monoxide molecules.

The results obtained by us show that the thermomagnetic effect can be used to study the magnetic properties of monolayers of chemisorbed particles.

The authors are very grateful to A.I. Zakharov, Yu.A. Matveev, and A.G. Narmonev for an analysis of the surfaces used in our experiments, to Yu.A. Perlovich for an analysis of the crystal structure of the evaporated metal films, and to Yu.M. Kagan, L.A. Maksimov, and S.Yu. Davydov for valuable discussions.

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Translated by A. Tybulewicz

Nuclear magnetic resonance of ^{51}V in a V$_3$Si single crystal above and below the structural transition temperature

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(Submitted 6 December 1978)
Zh. Eksp. Teor. Fiz. 77, 2313–2323 (December 1979)

We investigated the temperature dependences of the Knight shift, of the quadrupole-interaction constant, and of the spin-lattice and spin-spin relaxation times of ^{51}V nuclei, and also of the magnetic susceptibility in single-crystal V$_2$Si in the normal state. All the measured quantities with the exception of the spin-lattice relaxation time have singularities near the structural phase-transition temperature $T_M = 21$ K. As T_M is approached from above, a pretransition broadening of the quadrupole satellites is observed and is attributed by us to statistical distortions of the lattice near the defects. The hyperfine magnetic fields at the ^{51}V nuclei and the contributions of the wave functions of various symmetry to the density of the electronic states on the Fermi level are determined from the experimental data. The main contribution to the state density on the Fermi level is made by π-symmetry states.

PACS numbers: 76.60.Cq, 76.60.Es, 71.20.+c, 75.30.Cr

The intermetallide V$_3$Si, which has a crystal structure of the A-15 type, has unusual electron and phonon properties.[1] Particular interest attaches to the existence in V$_3$Si of a structural cubic to tetragonal phase transition at the point of the martensitic transition $T_M \approx 21$ K. In a number of theoretical models proposed to explain the anomalous properties of compounds of the A-15 type[2–4] it is assumed that the Fermi level passes near the narrow peak of the density of the electronic states, and the structural transition is attributed to instability of the electronic structure. At the present time, however, experimental data exist and make it possible to determine uniquely the details of the band structure of these compounds. It is therefore of interest to perform experiments that yield information on the electronic structure of V$_3$Si and on the character of its variation in phase transitions. This information can be obtained with the aid of nuclear magnetic resonance (NMR).

The NMR parameters are most sensitive to changes of the density of the electronic states on the Fermi level $N(E_F)$ and the symmetry of the nearest environment of the investigated nuclei. Direct information on the variation of $N(E_F)$ can be obtained also from measurements of the magnetic susceptibility χ, the results of which supplement the NMR data. A maximum of χ is observed at the temperature of the structural transition in V$_3$Si single crystals.[5] A splitting of the spectral