

TRANSITION OF A TWO-DIMENSIONAL PARAMAGNET INTO A MAGNETICALLY ORDERED STATE

Yu.S. Karimov

Institute of Chemical Physics, USSR Academy of Sciences

Submitted 8 February 1972

ZhETF Pis. Red. 15, No. 6, 332 - 336 (20 March 1972)

Phase transitions in layered structures of NiCl₂ and CoCl₂ with graphite were observed earlier [1]. Since such transitions in two-dimensional systems were very little investigated, it was of interest to study this phenomenon further. A second-order phase transition can be described, in accordance with the scaling hypothesis, by the set of critical indices [2]

$$C \sim |\tau|^{-\alpha}; \quad m \sim |\tau|^\beta; \quad \chi \sim |\tau|^{-\gamma};$$

$$m \sim H^{1/\delta}, \quad (T = T_c); \quad \chi \sim H^{-\lambda}, \quad (T = T_c);$$

where $\tau = (T - T_c)/T_c$ is the reduced temperature, H the magnetic field, C the specific heat, m the magnetization, and χ the magnetic susceptibility. The critical indices $\alpha, \beta, \gamma, \delta,$ and λ are connected by definite relations, so that specification of two indices and of the dimensionality of the space determines uniquely the entire set.

We have investigated NiCl₂ introduced into oriented pyrolytic graphite, the NiCl₂ layers being separated by not less than two layers of graphite. The interaction between neighboring magnetic layers is unusually weak [3], and the width of the electron-resonance line at room temperature shows that the anisotropy of the g-factor is small, $\Delta g/g < 0.01$. Its properties can therefore be described by starting from the two-dimensional Heisenberg model. A phase transition is observed at 18.1°K [1]. Figure 1 shows a plot of the reciprocal susceptibility against the square of the reduced temperature. We see that $\chi^{-1} = a\tau^2$ at small τ , i.e., $\gamma = 2$ in the paramagnetic region. Another index pertaining to the paramagnetic phase may be λ . It follows from Fig. 2 that in strong fields the magnetic moment can be represented in the form

$$m = m_0 + b \lg H$$

whence

$$\chi = \frac{\partial m}{\partial H} = b H^{-1}, \quad \lambda = 1.$$

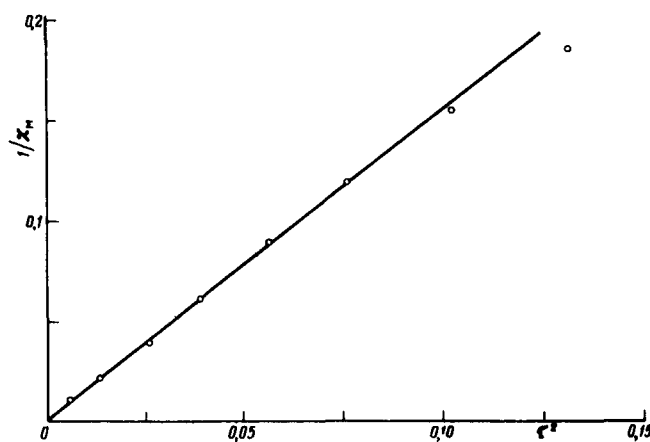


Fig. 1

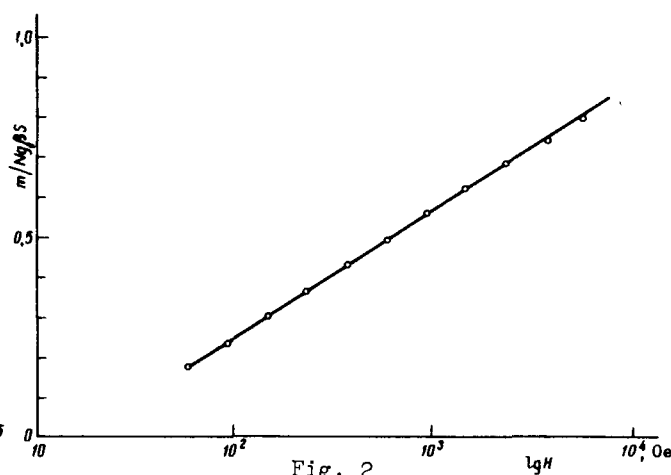


Fig. 2

Fig. 1. Reciprocal molar susceptibility (per g-mole of NiCl₂) vs. τ^2 .

Fig. 2. Dependence of the relative magnetic moment on $\lg H$.

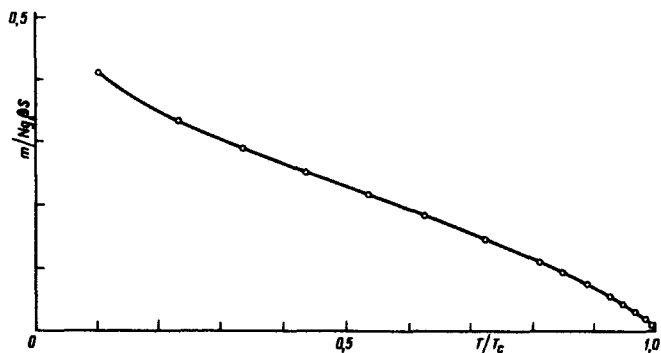


Fig. 3. Temperature dependence of the spontaneous ferromagnetic moment.

These values of λ and γ determine the set of indices for the paramagnetic phase:

$$\alpha = 0; \quad \beta = 0; \quad \gamma = 2; \\ \delta = \infty; \quad \lambda = 1. \quad (1)$$

Below 18.1°K the substance goes over into a magnetically ordered state, and the transition from this state into the paramagnetic state is characterized by an entirely different set of indices. At $T = T_c$ the $m(H)$ dependence in weak fields (1 - 100 Oe) along the direction of the

easy magnetization parallel to the layers is given by

$$m(H) = cH^{0.56 \pm 0.01}$$

whence $\delta' = 1.80 \pm 0.03$. No hysteresis is observed when the combination of NiCl_2 with graphite is magnetized in fields $H > 100$ Oe [1]. It was difficult earlier to obtain the complete magnetization curves in weaker fields, because of the use of a superconducting solenoid [1] that produced a residual field of opposite sign. Replacement of the superconducting solenoid by a copper one has made it possible to eliminate this effect. It then became clear that when the magnetizing field is removed a residual ferromagnetic moment is produced in the easy-magnetization plane. This proves that the given substance is in a magnetically ordered state. In the temperature interval $0.01 < |\tau| < 0.7$, the value of the spontaneous ferromagnetic moment (Fig. 3) can be written in the form

$$m = d|\tau|^{0.75 \pm 0.03}.$$

We thus have in this case the following set of critical indices:

$$\alpha' = -0.1 \pm 0.1; \quad \beta' = 0.75 \pm 0.03; \quad \gamma' = 0.63 \pm 0.05; \quad \delta' = 1.80 \pm 0.03. \quad (2)$$

It should be noted that the obtained values of β' , γ' , and δ' satisfy the relation [2]

$$\beta'\delta' = \beta' + \gamma'.$$

From an analysis of the critical indices (1) and (2) we can obtain information on the nature of the phase transition in a two-dimensional paramagnet. At $T > T_c$ the properties of the investigated compound are determined completely by the Heisenberg isotropic model. This is confirmed by the theoretical analysis of the model. Stanley and Kaplan [4] have proved the presence of a phase transition, while Lines [5] obtained the result $\gamma = 2$ for the Heisenberg two-dimensional model. At $T \leq T_c$ this model has very curious properties. First, from the fact that $\beta = 0$ it follows that there is no spontaneous magnetization. For isotropic two-dimensional systems this result was proved rigorously by Mermin and Wagner [6]. Second, $\delta = \infty$ corresponds to an infinitely large polarizability of the system. This makes the system unstable against small perturbations. Such perturbations are always present in real paramagnets. They may take the form of a weak interaction between neighboring layers, anisotropy of the g factor, or dipole-dipole interaction of the magnetic ions inside the layer. These interactions cause the paramagnet to go over into a magnetically-ordered state. The transition from this state into the paramagnetic state is characterized by the set of indices (2). The nature of the magnetically-

ordered state has not been finally established. This cannot be the usual ferromagnetic state, since the relative spontaneous moment does not exceed 0.5 (Fig. 3). It is most probable that the arrangement of the spins corresponds to a ferromagnetic helix with variable pitch.

- [1] Yu.S. Karimov, M.E. Vol'pin, and Yu.N. Novikov, ZhETF Pis. Red. 14, 217 (1971) [JETP Lett. 14, 142 (1971)].
- [2] V.L. Pokrovskii, Usp. Fiz. Nauk 94, 127 (1968) [Sov. Phys.-Usp. 11, 66 (1968)].
- [3] Yu.S. Karimov, A.V. Zvarykina, and Yu.N. Novikov, Fiz. Tverd. Tela 13, 2836 (1971) [Sov. Phys.-Solid State 13, No. 10 (1972)].
- [4] H.E. Stanley and T.A. Kaplan, Phys. Rev. Lett. 17, 913 (1966).
- [5] M.E. Lines, Phys. Rev. B., Solid State 3, 1749 (1971).
- [6] N.D. Mermin and H. Wagner, Phys. Rev. Lett. 17, 1133 (1966).

SELECTIVE LASER EXCITATION OF HIGH VIBRATIONAL LEVELS OF THE HCl MOLECULE

R.V. Ambartsumyan, V.M. Apatin, and V.S. Letokhov

Spectroscopy Institute, USSR Academy of Sciences

Submitted 11 February 1972

ZhETF Pis. Red. 15, No. 6, 336 - 339 (20 March 1972)

1. The most important problem in the problem of applying laser radiation selectively to a substance is direct selective excitation of the molecule vibrational levels. So far, only the first vibrational level of the molecule has been excited with laser radiation, and the higher levels were populated by collisions between the excited molecules. Unfortunately, molecule collisions lead to a loss of the selectivity of the excitation. We report here the first successful experiment on direct selective excitation of the third vibrational level of the HCl molecule ($E_{exc} = 1.04$ eV) by absorption of laser radiation at the second vibrational overtone of the molecule. The exceedingly high selectivity of such a process has made it possible to excite molecules having a definite isotopic composition.

2. The experiment consisted of selective optical pumping of the HCl molecule in the vibrational band $v = 0 \rightarrow v = 3$, using a high-power laser that was tunable in the IR region and detecting the excited molecules by means of IR luminescence of the vibrational transitions $v = 3 \rightarrow v = 2$ and $v = 2 \rightarrow v = 1$. The experimental setup is shown in Fig. 1. To excite the HCl molecule to the $v = 3$ level it is necessary to have radiation in the 1.18μ region with a frequency adjustable in a range $100 - 200 \text{ cm}^{-1}$. To this end, a special laser set-up was constructed using a driving laser with a tunable amplifier and stimulated Raman scattering (SRS) frequency converter. The neodymium laser with

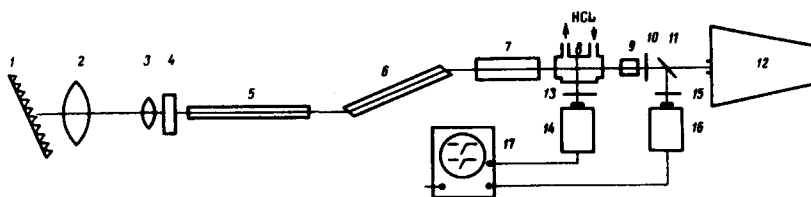


Fig. 1. Experimental setup: 1) diffraction grating, 2 - 3) telescope, 4) passive Q-switch, 5, 6) Nd rods, 7) cell with C₆H₅N, 8) cell with HCl, 9) KDP crystal, 10, 13, 15) filters, 11) beam-splitting plate, 12) STE-1 spectrograph, 14) Ge:Au receiver, 16) photomultiplier, 17) oscilloscope.