LARGE FLUCTUATIONS OF MAGNETIC ANISOTROPY OF CHROMIUM

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Submitted 19 June 1970
ZhETF Pis. Red. 12, No. 3, 137 - 139 (5 August 1970)

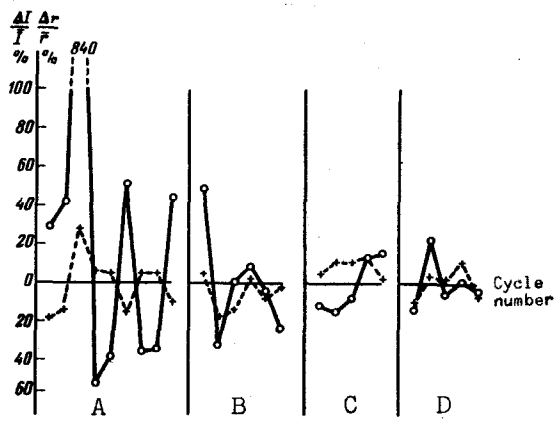
In the study of the magnetic structure of chromium, as shown in [1], it is necessary to take into account all the magnetic satellites of the sites of type (100), owing to their strong non-equivalence and owing to the presence of temperature hysteresis in the intensity of each satellite, which vanishes in the total sum. Werner, Arrott, and Kendrick [2], confirming the absence of hysteresis of the total intensity, observed a change in the temperature dependence of the satellites from experiment to experiment (within 30%). A similar influence was observed also by us. In addition, in certain samples we observed spontaneous changes of the intensities of the satellites at room temperature. This also indicates a possible instability of the magnetic structure of chromium.

In this study, we have attempted to reveal, with the aid of thermal cycling and magnetic working of the samples, the character of the instability of the magnetic structure and the influence exerted on it by the stability of the impurities. The investigation was carried out by a neutron diffraction method on five iodide single crystals of chromium (volume 60 - 70 mm^3). The first two samples contained 0.04 at.% of impurities (T_N = 310°K, T_{SF} = 125°K). The next three, judging from the values of the points T_N and T_{SF}, were less pure (sample No. 3 - 305 and 98°K, sample No. 4 - 314 and 106°K, sample No. 5 - 287 and 103°K, respectively). The reflection-measurement accuracy was 8%.

The thermal cycle consists of 10 min of annealing the sample above or below T_N, cooling in a jet of liquid nitrogen or of its vapor, and soaking for not more than 2 hours at 145°K, during which time we measured the intensities of the satellites (±b, 1, 0) and (1, ±b, 0) belonging to different modulation states. The ratio of these intensities will be denoted below by r, and their sum by I. The value of I was determined by averaging over all cycles.

In samples 1 and 2, which were purest according to our estimate, the thermal cycles caused the ratio r to and sum I to experience large fluctuations. Characteristic results for sample No. 1 are shown in the figure. We see that the largest fluctuations were experienced here by r, which revealed a tendency toward a considerable decrease with increasing lower limit of the cycles. Upon repetition of a series of cycles 300 - 143°K, in which the first cycle started with 343°K, we observed a memory effect: in each series r differed little from the value of r fixed upon passage through T_N. A change of the rate of passage through T_N from 1 deg/min to 1200 deg/min did not influence the character of the fluctuation.

The stability of the less pure samples No. 3 and No. 4 was not affected by the thermal cycles: No. 3 withstood twelve 343 - 143°K cycles without fluctuations, and No. 4 withstood sixty such cycles and in addition thirty cycles of 373 - 77°K. Sample 4 was then subjected, over a period of a month and a half, to more than sixty applications, at room temperature, of a magnetic field up to 20 kOe along the [100] direction, followed by annealing above T_N. The stability of the magnetic structure was monitored in the absence of a field by means of the peak (±b, 1, 0) at room temperature after preliminary annealing above T_N. As a result there appeared large (up to 40%) and apparently spontaneous fluctuations of the intensity in the intervals between the interactions of the field. Sample No. 5 was not made stable to the thermal cycles by the impurities: in seven out of 54 cycles (343 - 143°K) there were registered 18% fluctuations.



Percentage change of the ratio of the intensities of the reflections (±b, 1, 0) and (1, ±b, 0), belonging to different modulation states ($\Delta r/r$ - solid line) and also of their sum ($\Delta I/I$ - dashed line) in thermal cycling of the sample (T_N = 310°K, T_{SF} = 125°K). The series correspond to the following limits of thermal cycles and average values of r: a) 343 - 143°K, r = 1.46; b) 343 - 77°K, r = 3.10; c) 300 - 143°K, r = 1.02; d) 300 - 77°K, r = 5.25.

It is appropriate to report here also an unusual behavior of the single crystal investigated in detail in [1], which had a stable and distinctly pronounced magnetic anisotropy. After being left untouched for two years, it was observed that its anisotropy vanished. Measurements confirming this fact were carried out with a cryostat at nitrogen temperature and 1×10^{-6} mm Hg. Before these tests, as many times before, the sample was heated to 200°C. During the succeeding 15 runs in air and in nitrogen vapor, the distribution of the intensity in this crystal remained unchanged. Then the previous anisotropy was restored jumpwise, and remained unchanged during the course of the subsequent measurements in vacuum.

From an analysis of the experimental results it follows that:

1. Large changes of the magnetic anisotropy can occur in single-crystal chromium both under the influence of thermal or magnetic cycling, and spontaneously without special interaction (if one disregards small oscillations of 2 - 3° in the room temperature).

2. The absence of directivity, and the arbitrary changes accompanying identical cycles, make it possible to classify these changes as fluctuations. Increasing the upper point of the cycle above the Neel temperature contributes to a growth of the fluctuation amplitude, and a decrease of the lower temperature limit of the cycle causes the magnetic anisotropy to exhibit a tendency towards a directional growth.

3. The results of the investigation of samples 3 - 5 suggests that the thermal stability of the magnetic structure of chromium is more readily determined by the composition of the impurities rather than by their amount.

4. Each sample has apparently a dominating character of development of magnetic anisotropy.

If it is assumed that the proportions of the magnetic structure of chromium are due to mechanical stresses in the lattice of the sample, which are removed by annealing at 1000 - 1200°C [3], then our data indicate that these stresses exhibit certain singularities. First, they can be "relieved" without high-temperature annealing and return to the previous form, i.e., the general outline of the stress field in the sample can have a number of preferred configurations. Second, the stress field is sensitive to the temperature and can fluctuate when the latter varies. Third, it is sensitive to changes of the magnetic structure itself and can fluctuate under the influence of variations of this structure induced by direct action of an external magnetic field.

[1] V.S. Golovkin, V.N. Bykov, and V.A. Levdik, Zh. Eksp. Teor. Fiz. 49, 10 (1965) [Sov. Phys.-JETP 22, 6 (1966)].
[2] S.A. Werner, A. Arrott, and H. Kendrick, Phys. Rev. 155, 528 (1967).

[3] I. Bastow and R. Street, Phys. Rev. 141, 510 (1966).

DIRECT OBSERVATION OF HYPERBOLIC EXCITONS

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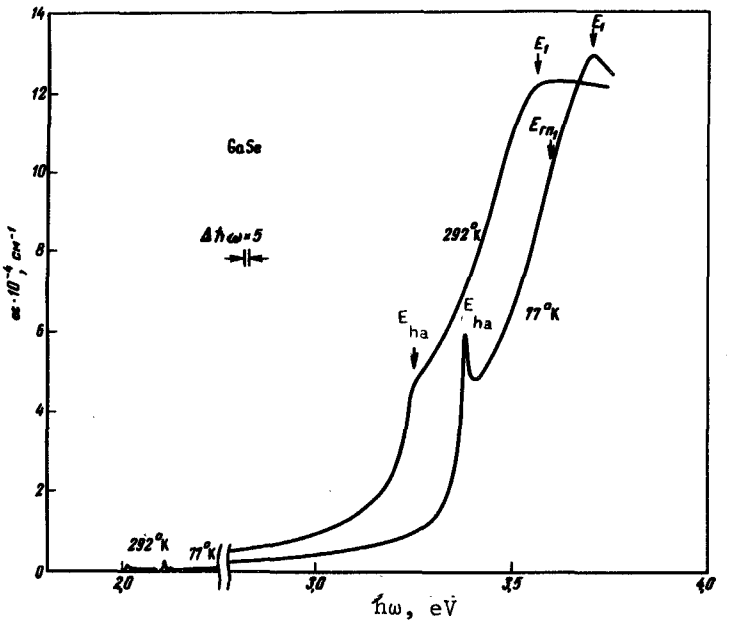
Submitted 22 June 1970
ZhETF Pis. Red. 12, No. 3, 139 - 141 (5 August 1970)

Phillips [1] proposed that electrons and holes can be bound into excitons not only at energies close to the energies of the absolute extrema, but also near the saddle points M_1 and M_2. Phillips called such excitons saddle-point excitons (hyperbolic excitons). He assumed that the existence of hyperbolic excitons can lead to the appearance of a distinct structure neat the points M_1 in the optical spectra. Velicky and Sak [2] and Kane [3] found that the Coulomb effects at the saddle critical points M_1 should become clearly pronounced in the optical spectra. Duke and Segall [4], on the other hand, theoretically refuted the existence of hyperbolic excitons. By measuring the transmission of thin samples of indium antimonide and cadmium telluride at low temperatures, Cardona and Harbeke [5] observed a structure which they ascribed to the exciton nature of the $L_3 \rightarrow L_1$ transition. Comparing the absorption coefficient above the energies of the first exciton peak with Elliott's formula [6] for allowed direct transitions, they obtained for the exciton binding energy values of 0.04 and 0.035 eV for InSb and CdTe, respectively. However, as indicated by Marple and Ehrenreich [7], another explanation of the structure is also possible. Shaklee, Rowe, and Cardona [8] confirmed, on the basis of the differential reflection spectrum of InSb near the critical point M_1, the existence of hyperbolic excitons in this material. Obviously the problem of the possible existence of bound states near saddle points and their manifestation in optical spectra is not yet ultimately solved.



Absorption spectra of GaSe above the edge of the fundamental absorption.

In this paper we present a clearcut proof favoring the existence of hyperbolic excitons with GaSe as an example.