New type of superstructures at the (100) surface of a cubic Heisenberg antiferromagnet EuTe

V. A. Grazhulis, A. M. Ionov, and V. F. Kuleshov
Institute of Solid State Physics, Academy of Sciences of the USSR

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The formation of a superstructure $(\sqrt{2}\times\sqrt{2})R-45°$, which is incompatible with the bulk spin structure, has been observed experimentally for the first time on a clean (100) surface of a cubic antiferromagnetic semiconductor EuTe at $T \lesssim T_N \sim 9.6$ K. The formation of this superstructure upon going through T_N from the high-temperature side occurs through the formation of an "intermediate" modulated superstructure which undergoes a transition to $(\sqrt{2}\times\sqrt{2})R-45°$ upon lowering the temperature. The wave vector **k** of a modulated superstructure changes reversibly as T is varied from the center, $\mathbf{k}=0$, to the edge of the square two-dimensional Brillouin zone, always remaining oriented along its diagonal.

The EuTe crystals fall within the category of ionic Heisenberg antiferromagnetic semiconductors with a NaCl cubic lattice. The Eu^{2+} ions are the magnetic ions, $S=7/2$. At $T<T_N$, where T_N is the Néel temperature (~ 9.6 K), an antiferromagnetic order with a spin space lattice, shown in Fig. 1a, appears in the bulk of the crystal.

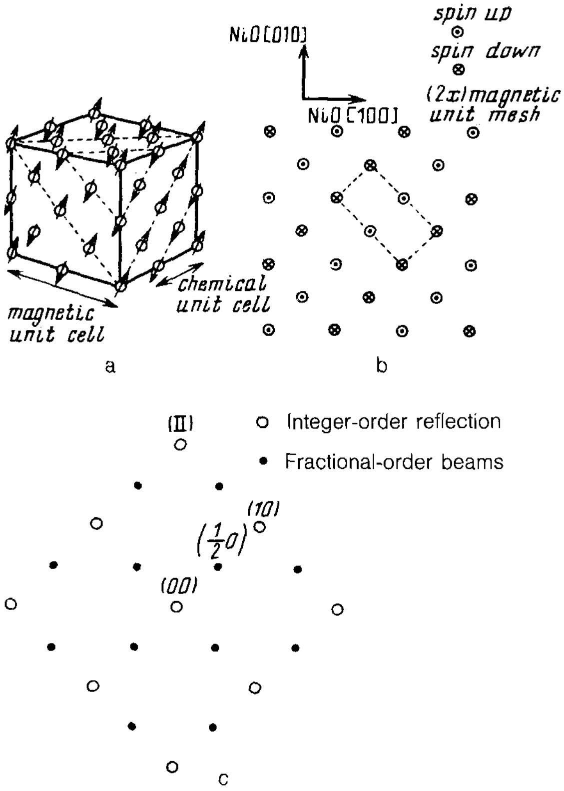

FIG. 1. Spin order and diffraction of low-energy electrons by NiO (from Ref. 2). a—Spin order of NiO; b—spin order at the (100) surface of NiO; c—diffraction of low-energy electrons from the (100) surface of NiO below T_N (diffraction occurs from several domains).

Using the low-energy electron-diffraction (LEED) method, we have previously shown[1] that the pure (100) cleavage faces of EuTe have an unreconstructed atomic structure (1×1) over the entire temperature interval $T = 10$–300 K. In the present letter we report the results of a further study of the (100) surfaces of EuTe at lower T, including $T_N \sim 9.6$ K, by the LEED method; specifically, in the interval 8–15 K. The only magnetic materials whose surface structure near T_N has so far been studied are, to the best of our knowledge, the crystals[2,3] NiO. At $T < T_N$ the crystals EuTe and NiO have identical lattices and identical spin structures. At $T < T_N \sim 520$ K the compound NiO (100) reveals the presence of weak superreflections in the low-energy electron-diffraction pattern (Fig. 1), indicating the onset of spin order at the (100) surface (Fig. 1b), which is a consequence of a bulk antiferromagnetic order (Fig. 1a).

In our experiments we studied the n-type EuTe crystals, $\rho \sim 5\ \Omega\cdot$cm, which were grown by the gas-transport method with an excess of Eu. The typical dimensions of the crystals were $1.5\times2\times5$ mm. The crystals were cleaved in the (100) plane. The LEED patterns were observed in $\sim10^{-10}$-torr vacuum. For this purpose we used low-temperature sample holders which were designed and built by us previously (the crystal, held in the sample holder by tightening the copper screws, was clamped close to the [100] direction).

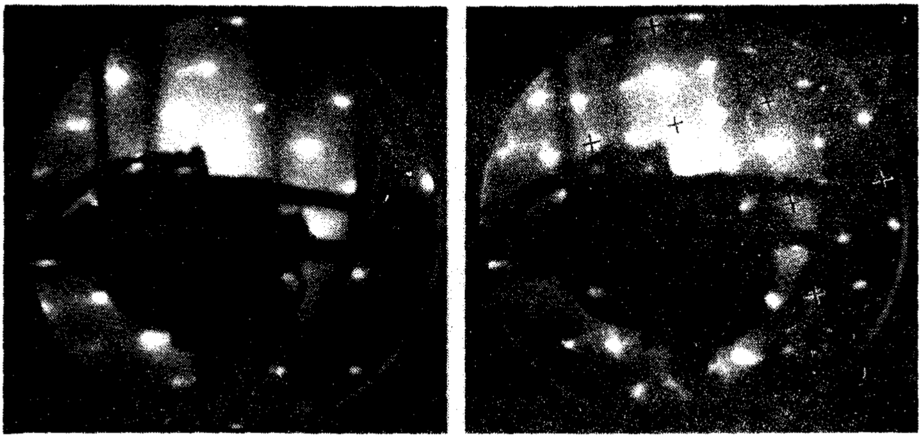

FIG. 2. Diffraction of low-energy electrons from the (100) surface of EuTe. a—$T = 15$ K; b—$T \sim 8$ K (several superreflections are marked by crosses).

After the EuTe crystals are cleaved, we observe at 15 K a clearly defined LEED pattern (Fig. 2a) corresponding to a nonreconstructed[1] EuTe (100) surface, (1×1). As T is lowered, we see a curious phenomenon: the appearance of superreflections which initially split off from the principal reflections and which move along the direction of one of the diagonals toward the centers of the squares as the temperature is lowered (Fig. 2b). The superreflections are seen in the interval $E_p = 80$–140 eV and the principal reflections are seen in the interval 80–650 eV, where E_p is the energy of the primary electron beam. At $T \sim 8$ K all superreflections are localized at the centers of the squares. We ultimately see the formation of a LEED pattern, shown in Fig. 2b. A multiple scanning of the temperature over the interval 8–15 K shows that the changes observed in the LEED pattern are totally reversible.

As in the experiments[3] on NiO, we were able to observe superreflections only in some of the crystals, specifically, in only one of the three EuTe crystals that were tested. The LEED pattern with superreflections was clearly observed in $\sim 10^{-10}$-torr vacuum for several hours. The subsequent heating of the sample to $T \sim 300$ K in ~ 40 min led to an increase in the pressure to $\sim 5 \times 10^{-9}$ torr in the chamber, apparently causing some uncontrollable contamination of the cleaved surface. Superreflections were not seen at all when the temperature was lowered again (after warming the sample to ~ 300 K), although the principal reflections were clearly seen, as previously, at all temperatures. We can assume, therefore, that a highly pure and possibly crystallographically perfect cleaved (100) surface of EuTe will have superstructural reflections.

The wave vector $\mathbf{k}$, which characterizes the superstructure, runs through the values from $\mathbf{k} = 0$ to $\mathbf{k} = \mathbf{k}_0$ upon changing T, where $\mathbf{k}_0$ is a vector corresponding to the points X (the center of the Brillouin zone) (Fig. 3c). Clearly, the intermediate values of $\mathbf{k}$ correspond to the incommensurate superstructures. Notice that the low-temperature superstructure which we detected $(\sqrt{2} \times \sqrt{2})R - 45°$ (Fig. 3b) differs fun-

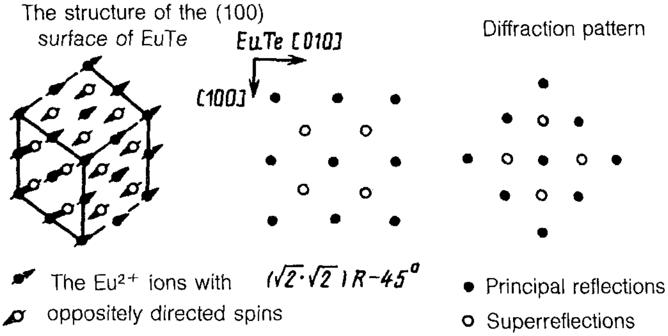

FIG. 3. Spin order at the (100) surface of EuTe. a,b—The structure of the (100) surface of EuTe (the Te^{-2} ions are not shown); c—diffraction of low-energy electrons from the (100) surface of EuTe at $T \sim 8$ K.

damentally from the expected superstructure (2×1), shown in Fig. 1b. In other words, at $T<T_N$ the (100) surface superstructure of EuTe which is formed does not match the volume structure, in contrast with the (100) surface of NiO. This result, along with the intermediate incommensurability, is a fundamentally new result.

There are grounds for assuming that the superreflections observed in the LEED pattern are due, just as in the case of the (100) surface of NiO, to the exchange scattering of a primary (spin-unpolarized) electron beam by a spin lattice which is formed at $T<T_N$. A certain atomic reconstruction of the surface that accompanies the magnetic ordering can, in principle, contribute to the intensity of superreflections, but the magnitude of this effect is so far unknown. The formation of an incommensurate superstructure which transforms to $(\sqrt{2}\times\sqrt{2})R-45°$ can be attributed, in our view, to the formation of an incommensurate helicoidal spin structure at $T<T_N$ near the (100) surface of EuTe. The EuTe crystal acquires order when the angle of flipping of the spins in the adjacent planes is equal to π, consistent with the antiferromagnetic spin order in Fig. 3a. Clearly, if the spins in the adjacent planes are flipped near the (100) surface by an arbitrary angle, a spin superstructure with an incommensurate periodicity will form at the surface along the [100] direction (Fig. 3a), in qualitative agreement with the experimental data obtained by us. Further experimental studies must be carried out in order to determine the precise mechanism responsible for the formation of surface superstructures that have been observed, when there are no such structures in the bulk of the crystal. In particular, the $\mathbf{k}(T)$ curve must be found and the nature of the phase transition at the EuTe surface near T_N must be determined.

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