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Superconducting Properties of the Atomically Disordered MgB_2 Compound

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The effect of disorder induced by neutron irradiation in a nuclear reactor (thermal neutron fluence 1×10^{19} cm^{-2}) on the superconducting transition temperature T_c and the upper critical field H_{c2} of polycrystalline MgB_2 samples was investigated. Despite the appreciable radiation-induced distortions (more than ten displacements per atom), the initial crystal structure (C32) was retained. The temperature T_c decreased from 38 to 5 K upon irradiation and was practically completely restored after the subsequent annealing at a temperature of 700°C. A weak change in the dH_{c2}/dT derivative upon irradiation is explained by the fact that the irradiated samples are described by the "pure" limit of the theory of disordered superconductors. The suppression of T_c upon disordering may be due to the isotropization of the originally anisotropic (or multicomponent) superconducting gap or to a decrease in the density of electronic states at the Fermi level. *© 2001 MAIK "Nauka/Interperiodica".*

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The discovery of superconductivity (SC) in binary MgB_2 compounds with $T_c \approx 40$ K [1] has raised the question of what the reason is for high T_c values and SC mechanism in such a simple system. Within the framework of electron–phonon superconductivity, the high T_c values are favored by high densities $N(E_F)$ of electronic states at the Fermi level and by soft phonon modes [2], although an exotic pairing mechanism may also take place in this particular case. The response of a system to a disorder induced by the irradiation with high-energy particles serves as a test for revealing the specific features of the SC state.

In metals (of the Nb type), the presence of several percent of impurity atoms or radiation-induced defects reduces T_c only slightly because of a small gap anisotropy. In the intermetallic compounds of the A15 type, a more substantial radiation-induced disorder may occur (suppression of long-range order and amorphization) because of considerable transformations in the electronic and phonon systems. In compounds with high $N(E_F)$ and T_c values (Nb_3Sn, V_3Si), the radiation-induced disordering lowers T_c (from 15–20 K to 1–3 K) because of a decrease in $N(E_F)$ [3]. In the compounds with low $N(E_F)$ and T_c (Mo_3Si and Mo_3Ge), the disordering increases T_c (from 1.5 to 7 K) because of an increase in $N(E_F)$ and softening of the phonon mode. This signifies that the individual features of electronic structure vanish upon the disordering, and the superconducting properties of the disordered compounds become similar to the properties of amorphous materials. At the same time, the effect of disorder on the SC of high-T_c superconductors radically differs from that observed in A15; the irradiation rapidly destroys SC ($T_c = 0$) [4].

In this work, we studied the superconducting properties of polycrystalline MgB_2 samples ($0.05 \times 1 \times 5$ mm) irradiated in the core of an IVV-2M nuclear reactor (thermal- and fast-neutron fluences $\Phi = 1 \times 10^{19}$ and 5×10^{18} cm^{-2}, respectively) or obtained after the subsequent isochronal annealing for 20 min at temperatures $T_{ann} = (200–700)$°C. MgB_2 powders were prepared by the method described in [5] and pressed under a pressure of 9 GPa at room temperature without the subsequent thermal treatments. According to X-ray powder diffraction analysis (CuK_α radiation), the samples contained traces (~3%) of MgO. This method of sample preparation gave a strained structure and was accompanied by the broadening of X-ray reflections and smearing of the superconducting transitions in the resistivity $\rho(T)$ and ac susceptibility $\chi(T)$ curves (Fig. 1), as well as by an increase in the resistivity $\rho(T)$ ($\rho \approx 0.4$ mΩ cm at $T = 300$ K). The latter fact implies that the intergrain boundaries make a considerable contribution to ρ(T), so that we will not analyze our data using the resistivity curves.

The structural distortions induced by the irradiation of B-containing materials by thermal neutrons are mainly due to the nuclear reaction at the ^{10}B isotope with emission of α particles and production of ^{7}Li. At $\Phi = 1 \times 10^{19}$ cm^{-2}, almost 1% of B atoms are splitted, which corresponds to a damaging dose of approximately more than 10 displacements per atom. Although there is a considerable absorption of thermal neutrons in the sample, one can expect that, due to multiple displacements of each atom, the radiation-induced defects are homogeneously distributed over the sample volume. In spite of the fact that the radiation-induced distortions of the material are very strong, the symmetry of the original structure (C32) is retained after the irradiation. The structural parameters refined by using the Rietveld analysis are given in the table. The irradiation resulted in the anisotropic expansion of the crystal lattice (the unit-cell volume increased by 1.4%), a faster increase in the c parameter, and a decrease in the occupancy N(Mg) of magnesium sites [N(B) = 1].

Figure 1 demonstrates the transformations of superconducting transition curves for the resistivity and susceptibility of the irradiated and annealed samples. The radiation reduces T_c down to ≈5 K, while the annealing at 700°C almost completely restores T_c. The transition width changes only slightly upon the irradiation, in compliance with the assumption about the homogeneous distribution of defects. The ρ(T) and χ(T) curves of the initial sample have a characteristic "two-step" shape, which is retained after the irradiation and the annealing below T_{ann} = 500°C (Fig. 1). It is likely that the low-temperature step is due to the presence of a strongly strained structure with lower T_c in the regions adjacent to the grain boundaries. For T_{ann} = (600–700)°C, this step smears, probably, due to the loss of Mg at the grain surfaces. The resistive transitions do not show any noticeable broadening in a magnetic field (Fig. 2). The temperature curves for the upper critical field H_{c2}, as determined from the half-transition temperatures (0.5 of the normal-state resistivity ρ_n), have almost identical slopes for the initial and irradiated samples (Fig. 3).

As was pointed out above, a considerable contribution to the resistivity of the samples studied comes from the intergrain boundaries; its true value for the MgB_2 grains is unknown. Experiments [6] with dense MgB_2 "wires" show that the mean free path l_{tr} in them is as high as 600 Å, whereas the coherence length ξ_0, as determined from the H_{c2} measurements, is on the order of 50 Å. For this reason, a weak change in the H_{c2}/dT derivative in our experiments can be explained by the fact that the irradiated samples remained in the region of a "pure" limit $l_{tr} > \xi_0$ (residual resistivity increased after the irradiation no more than eightfold). The theoretical calculation of dH_{c2}/dT in a simple model of a superconductor with anisotropic s pairing [7] indicates that the curve slope for the critical field changes only slightly in the whole region of transition from the

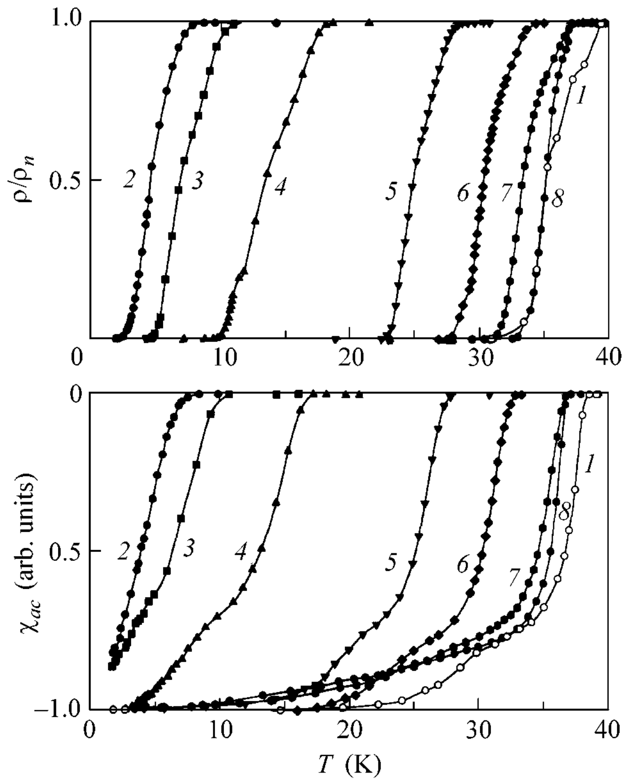

Fig. 1. Temperature dependences of the (up) reduced resistivity ρ/ρ_n and (down) the ac susceptibility χ_{ac} of the (*1*) initial MgB_2 samples, (*2*) samples irradiated by the thermal-neutron fluence $\Phi = 1 \times 10^{19}$ cm^{-2}, and (*3*–*8*) samples annealed at T = 200, 300, 400, 500, 600, and 700°C, respectively. $\rho_n = \rho(T > T_c)$; $\chi_{ac} = -1$ corresponds to the complete screening of a sample by the superconducting currents.

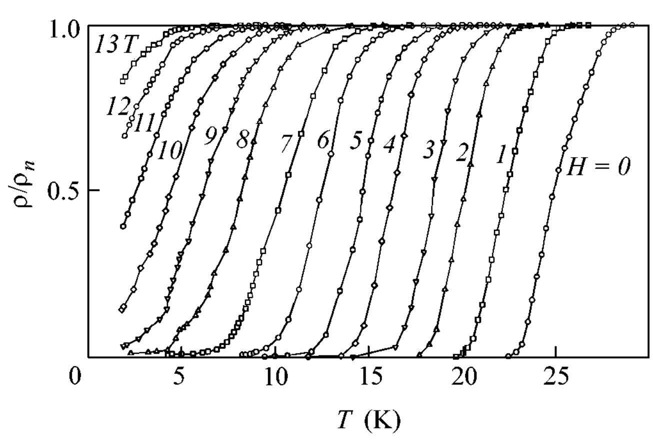

Fig. 2. Temperature dependences of ρ/ρ_n for the MgB_2 samples irradiated and annealed at T = 400°C in magnetic fields denoted by numbers in units of T.

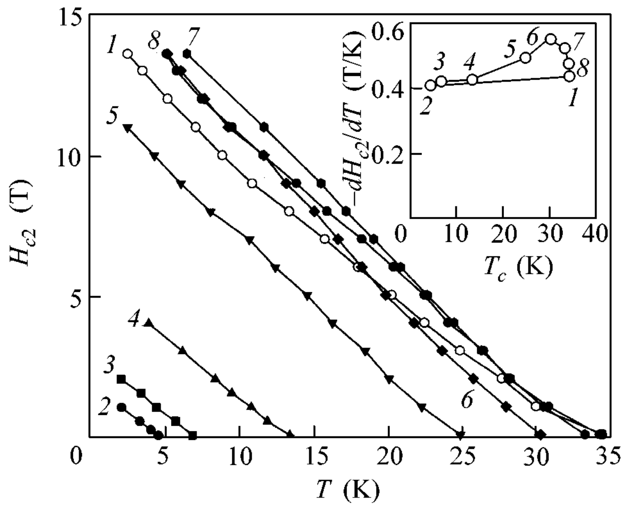

Fig. 3. Temperature dependences of the upper critical field H_{c2} for the initial, irradiated, and annealed MgB_2 samples. Inset: dependence of $(-dH_{c2}/dT)$ on T_c. Notations are as in Fig. 1.

"pure" to the "dirty" limit. At the same time, the T_c temperature noticeably decreased in this region because of the isotropization of the energy gap. Note that the behavior of T_c observed in [7] is not universal and depends on the anisotropic model of the initial gap. In principle, T_c can strongly decrease depending on the parameters of the initial anisotropy.

The strong lowering of T_c, observed in this work, can be even more justifiably assigned to a decrease in the density $N(E_F)$ of electronic states at the Fermi level upon the disordering. In the dirty limit, $l_{tr} < \xi_0$, the slope of the H_{c2} curve starts to increase with a decrease in the mean free path [7]. This effect was practically unobserved in our experiment, which may be explained both by the fact that the dirty limit was not attained and by the fact that the corresponding increase is masked by the lowering of $N(E_F)$ upon the disordering. Independent indications that the anisotropic pairing or a closely related SC with different superconducting gaps at different Fermi surface sheets MgB_2 (multicomponent gap) is possible were provided by the measurements of specific heat [8] and the temperature dependence of penetration depth [9], as well as by the calculations in [10].

Lattice parameters a and c, unit-cell volume V, and occupancies N(Mg) of magnesium sites for the initial MgB_2 samples and the samples irradiated by neutron fluence $\Phi = 1 \times 10^{19}$ cm^{-2}

	Initial	Irradiated
a, Å	3.0878(2)	3.0953(3)
c, Å	3.5216(4)	3.5533(4)
V, Å^3	29.080(4)	29.482(5)
c/a	1.140	1.148
N(Mg)	0.96(4)	0.89(5)

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