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CENTRAL RESEARCH INSTITUTE FOR PHYSICS

BUDAPEST

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АННОТАЦИЯ

Изучалось электрическое сопротивление и термическое напряжение аморфных систем Fe-B, Fe-B-Si, Fe-Co-B, Fe-Ni-B и $Fe_{80}TM_3B_{17}$ /где TM = один из 3d-, 4d- или 5d-переходных металлов/ от комнатной температуры до температуры аморфно-кристаллического перехода. Для интерпретации экспериментальных результатов использовалась видоизмененная "расширенная теория Займана", разработанная для неупорядоченных систем.

KIVONAT

A Fe-B, Fe-B-Si, Fe-Co-B, Fe-Ni-B és $Fe_{80}TM_3B_{17}$ (ahol TM a 3d, 4, vagy 5d átmeneti fémek valamelyike) amorf rendszerek elektromos ellenállását és termofeszültségét mértük szobahőmérséklettől az amorf-kristályos átalakulásig. A kisérleti eredmények értelmezése a rendezetlen rendszerekre kidolgozott un. "kiterjesztett Ziman-elmélet" módositása utján történt.

ABSTRACT

The electrical resistance and the thermopower of the Fe-B, Fe-B-Si, Fe-Co-B, Fe-Ni-B and $Fe_{80}TM_3B_{17}$ amorphous systems (the TM is one of the 3d, 4d or 5d transition metals) have been measured from room temperature to the amorphous-crystalline transformation. The extended Ziman theory for the electrical transport properties of structurally disordered systems was modified and that was used to interprete some of the experimental data.

INTRODUCTION

Recently there is a considerable growth of interest in the electrical transport properties of metallic glasses. A great number of experimental and theoretical investigations were published aiming to understand mainly the temperature dependence of these properties and relatively less work was done on the effect of composition change. Our aim was to study a group of materials based on the Fe-B system changing systematically both the metallic and the metalloid components. We tried to interprete the trends with composition in the amorphous state by using a modified version of the extended Ziman-Faber theory for the transport properties. By studying the crystallization we looked for a correlation between the alloying elements added to the Fe-B system and the resulting crystalline phases.

The electrical resistivity and the thermopower of $Fe_{100-x}B_x$, $Fe_{80}B_{20-x}Si_x$, $(Fe_{100-x}Co_x)_{75}B_{25}$, $(Fe_{100-x}Co_x)_{80}B_{20}$, $(Fe_{100-x}Ni_x)_{75}B_{25}$, $(Fe_{100-x}Ni_x)_{85}B_{15}$, $Fe_{80}TM_3B_{17}$ amorphous alloys were measured from room temperature to the

crytalline transition for different compositions. In the last group of samples TM stands for one of the 3d, 4d and 5d elements. The ribbons were made by quenching to the external surface of a fastly rotating copper disk and analyzed by atomic absorption spectrometry.

The classical four-probe method and a pure Al-sample differential thermocouple set-up were used for the measurement of the resistance and the thermopower, respectively. The measuring system is based on two Keithley nanovoltmeters, a Solartron Data Logger and a HP-97-S calculator.

RESULTS

The experimental data for the amorphous state and for the amorphous-crystalline transition are collected on Figs. 1-7. and Figs. 8-10., respectively. For different concentrations of a

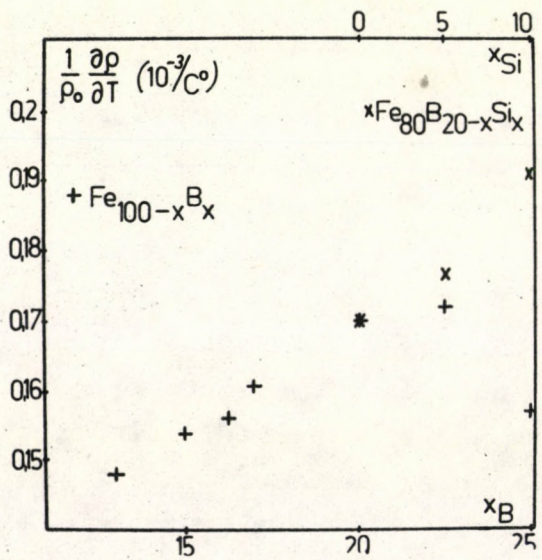


Fig. 1. Temperature coefficient of the electrical resistivity at 273 K for different compositions of Fe-B and Fe-B-Si

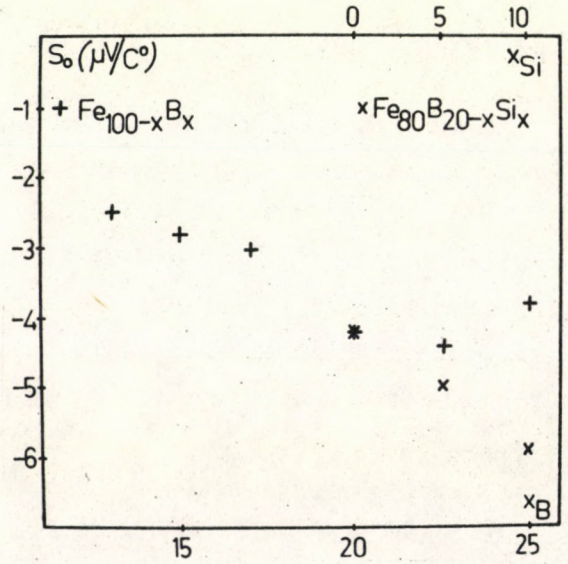


Fig. 2. Absolute termopower at 273 K for different compositions of Fe-B and Fe-B-Si

given system the transport properties are showing sometimes monotonous behaviour /e.g. for the Fe-Co-B and Fe-B-Si systems/,

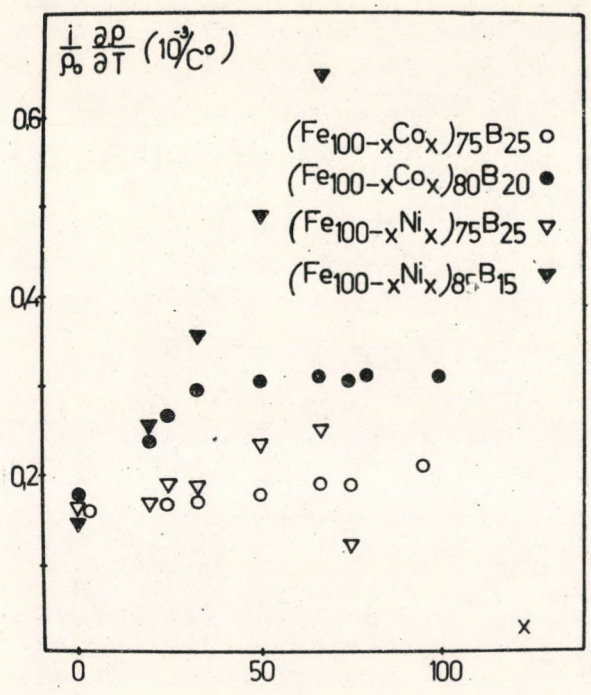


Fig. 3. Temperature coefficient of the electrical resistivity at 273 K for different composition of Fe-Co-B and Fe-Ni-B

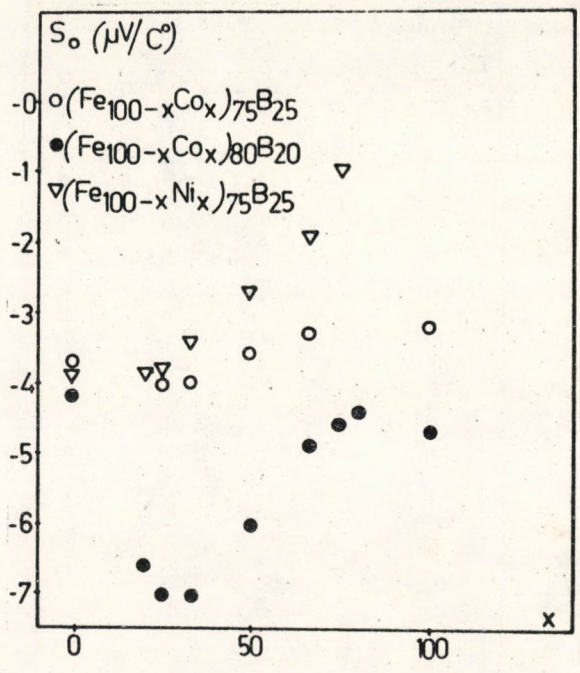


Fig. 4. Absolute thermopower at 273 K for different compositions of Fe-Co-B and Fe-Ni-B

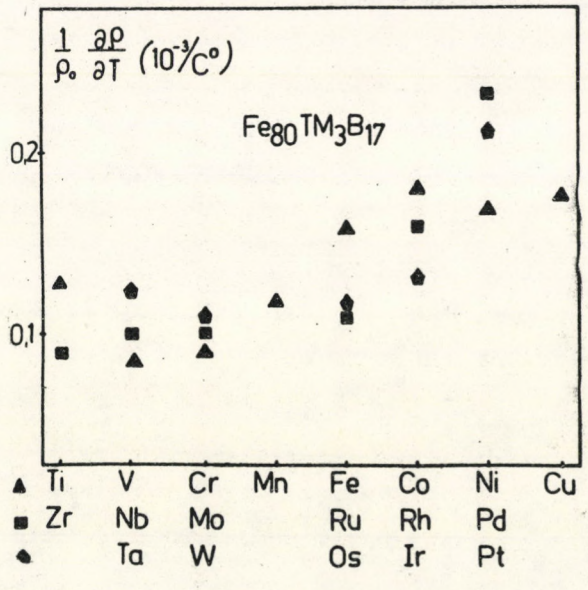


Fig. 5. Temperature coefficient of the electrical resistivity at 273 K for $Fe_{80}TM_3B_{17}$

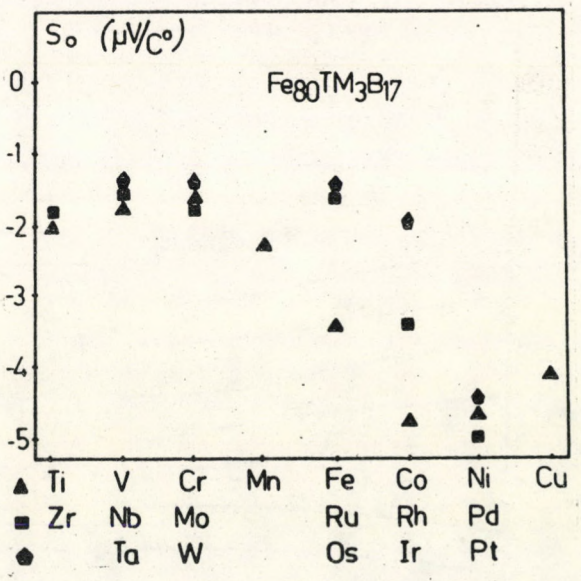


Fig. 6. Absolute thermopower at 273 K for $Fe_{80}TM_3B_{17}$

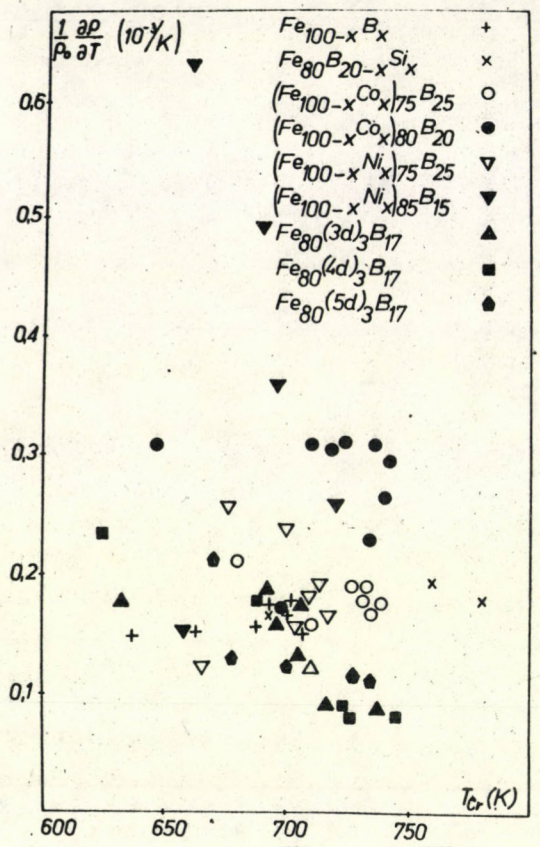
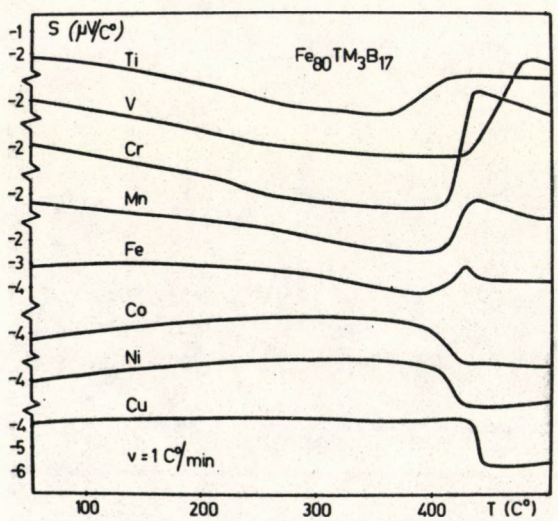


Fig. 7. Temperature coefficient of the electrical resistivity at 273 K for "Fe-B family" related to the crystallization temperature



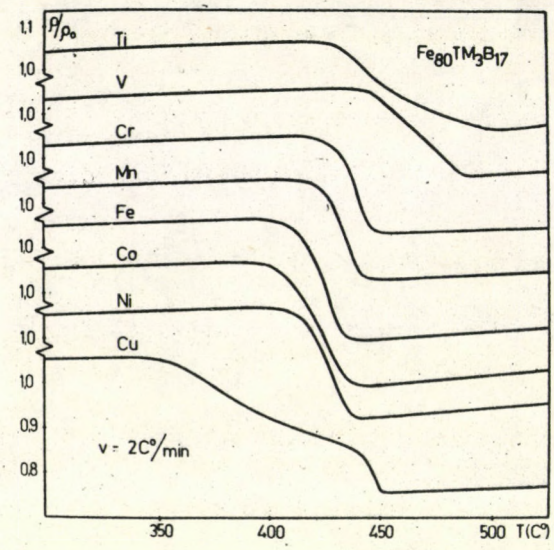


Fig. 8. Normalized electrical resistivity as a function of the temperature for samples of $Fe_{80}(3dTM)_3B_{17}$

in other cases a slight extremum occurs /e.g. for the Fe-B system/. If the temperature coefficient of the electrical resistivity /TCR/ increases or decreases as a function of the composition, generally the thermopower decreases or increases respectively. The collected data on Fig. 7. are not supporting the earlier suggestion that the lower the TCR the higher is the stability of the amorphous state. On the amorphous-crystalline transition the changes of the

Fig. 9. Absolute thermopower as a function of the temperature for samples of $Fe_{80}(3dTM)_3B_{17}$

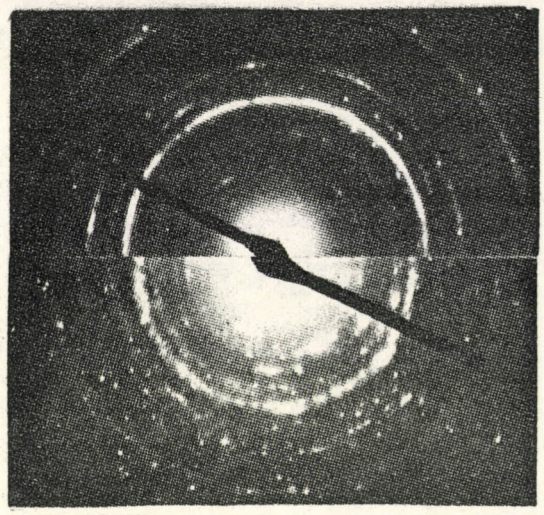


Fig. 10. Diffraction patterns for samples of the crystallized $Fe_{80}(3dTM)_3B_{17}$
a, Fe_2B phase is preferred when 3dTM is left to Fe
b, Fe_3B phase is preferred when 3dTM is right to Fe

transport properties are determined mainly by the crystallization products. According to the electron diffraction patterns Fe_2B phase is preferred (besides the alfa-Fe) by the samples having 3d alloying elements left to Fe, while the Fe_3B phase occurs in the samples with elements right to Fe. Preliminary measurements are showing the same tendency with 4d and 5d elements too.

The experimental data are used to check the validity of the extended Ziman-Faber theory [1] for the electrical transport properties of liquid and amorphous metals. The formulae for the resistivity and for the thermopower are:

$$\rho = (3\pi\Omega_o/\hbar e^2 v_f^2)(4k_f^4)^{-1} \int_o^{2k_f} dq\, q^3 |T|^2, \qquad (1)$$

$$S = -(\pi^2 k_B T/3|e|E_F)X_S, \qquad X_S = -[\partial \ell n\rho(E)/\partial \ell nE]_{E=E_F} \qquad (2)$$

where for a binary system

$$|T|^2 = c_1|t_1|^2[1-c_1+c_1a_{11}(q)]+c_2|t_2|^2[1-c_2+c_2a_{22}(q)] +$$

$$+ c_1c_2(t_1^x t_2+t_1 t_2^x)[a_{12}(q)-1]. \qquad (3)$$

Here t_1 and t_2 are the t-matrices of the individual atoms, $a_{ij}(q)$ are the partial structure factors.

The E_F Fermi energy is determined by solving the "generalized Friedel sum rule" for the energy [2]

$$c_1 Z_1 + c_2 Z_2 = (\Omega_o E^{3/2}/3\pi^2) + \\ + (2/\pi)\sum_{\ell}(2\ell+1)[c_1 \eta_\ell^1(E) + c_2 \eta_\ell^2(E)] + N_m(E) \equiv F(E). \qquad (4)$$

Here Z_1 and Z_2 are the total number of electrons per atom in the conduction band, $\eta_\ell^1(E)$, $\eta_\ell^2(E)$ are the scattering phase shifts as functions of energy for the atoms No1 and No2, respectively. $N_m(E)$ is a correction due to multiple scattering effects.

The input quantities for the calculations were

a, Herman-Skillman atomic wave functions,

b, the experimental structure factors, fitted by the partial structure factors, $a_{ij}(q)$, from the hard sphere solution of the Percus-Yevick equations, supposing the T-depence through a Debye-Waller factor, and

c, the experimental value for the density.

The method of calculations for binary systems started by fitting the parameters of $a_{ij}(q)$ - packing density, and ratio of the hard sphere diameters - to the experimental structure factor. This was followed by Fourier transforming the a_{ij}'s in order to get the partial distributions, $g_{ij}(R)$. Using the atomic wave functions and this $g_{ij}(R)$ a muffing-tin potential was constructed by the way proposed originally by Mattheiss [3] and applied for liquid metals by Mukhopadhyay et al. [4]. The solution of the radial Schrödinger equation was used to calculate the phase shifts and their derivatives, as a function of energy. After solving equation (4) for the Fermi energy the (1) integral and equations in (2) gave the results for the resistivity and thermopower. Repeating the calculations for higher temperatures we calculated the TCR above room temperature.

Ternary systems were treated as quasi-binary system by assuming that the structure remains the same when a binary system is alloyed, and also the scattering properties - the phase shifts, as a function of energy - are the same for all the transition elements as in the corresponding binary system, having the same

concentration of transition-metal, metalloid atoms. Only the density and the electron/atom ratio was changed compared to the binary system with the same c_1,c_2 giving rise to new values of Fermi energy, E_F, to new phase shifts and to derivatives.

A typical data set and results are shown in Table 1., the applied functions can be seen on Figs. 11-12. Calculations were performed for all samples for which experimental measurements were carried out. By analysing the results in comparison with experimental data one can conclude as follows:

The Fermi energy, E_F, turns out to be relatively high /measured from the muffin-tin zero/, which corresponds to a broad conduction band. The position of the first peak of S(q), q_1, is lower than $2k_F$ for all the materials used, in agreement with the positive value of TCR. When one neglects the only undefined

Table 1. A typical data set for amorphous $Fe_{83}B_{17}$

Input data:	packing density	η=0.55
	ratio of hard sphere diameters	α=0.698
	experimental value of density	ρ_o=7.47 g/cm^3
	Debye temperature	Θ_D=200 K
	temperature	T=300 K
Calculated data:	hard sphere diameter of Fe	S_{Fe}=4.406 au.
	hard sphere diameter of B	S_B=3.077 au.
	atomic volume	Ω_o=72.29 au.
	average Wigner-Seitz radius	R_{WS}=2.584 au.
	number of electrons/atom	Z=7.15
	multiple scattering correction	N_m=-1.9
	Fermi energy	E_F=1.107 ry
	Fermi wave number	$2k_F$=3.98 A^{-1}
	phase shifts of Fe at E_F	η_o=-0.782
		η_1=-0.182
		η_2=2.506
	phase shifts of B at E_F	η_o=0.303
		η_1=0.818
		η_2=0.018
	electrical resistivity /T=300 K/	
	calculated value	ρ_{calc}=129.5 $\mu\Omega$cm
	experimental value	ρ_{exp}=133 "
	temperature coefficient of ρ	
	calculated value	TCR_{calc}=2.34 10^{-4}/K
	experimental value	TCR_{exp}=1.61 "
	thermopower /T=300 K/	
	calculated value	S_{calc}=-2.6 μV/K
	experimental value	S_{exp}=-3.1 "

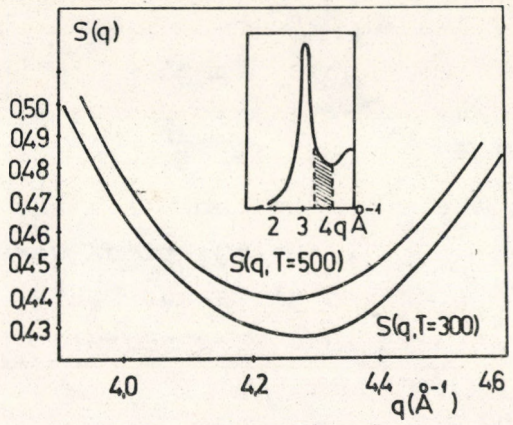


Fig. 11. Temperature-dependent structure factors for $Fe_{83}B_{17}$ from Percus-Y. hard spheres. Insert shows where the $2k_F$ values (shaded area) of our samples are, compared with the first peak of S(q)

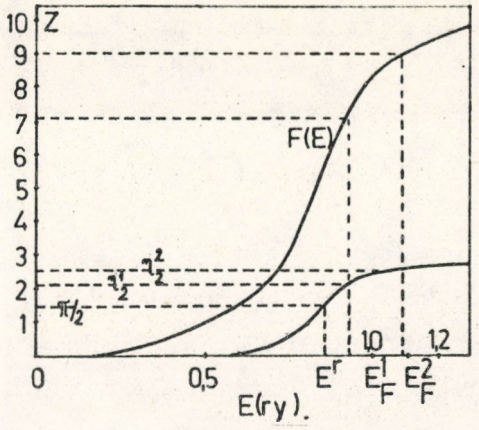


Fig. 12. The right hand side, F(E), of the "generalized Friedel sum rule" and the d-phase shift, $\eta_2(E)$, of Fe atoms in $Fe_{83}B_{17}$.
E^r: the resonant energy, E_F^1 and E_F^2: Fermi energy without and with N_m

parameter, the multiple scattering correction, $N_m(E)$, then too small values for $\eta_2^{Fe}(E_F)$, consequently, too large values for the resistivies are resulted, as in earlier calculations /Esposito et al. [2]/. If one fits $N_m(E)$ to the experimental value of the resistivity for a binary alloy of a given concentration /to $Fe_{83}B_{17}$ in our case/, then one gets reasonable results for other samples, including ternary systems, too, using the same value of $N_m(E)$. Since $2k_F$ is above, and relatively far from q_1, the tendencies with changing the concentrations and compositions of the samples are rarely reproduced by this simplified treatment, using only the density and the number of electrons as parameters.

To summarize, one can say that the extended Ziman-theory gives qualitatively good results when applied rigorously, taking into account the multiple scattering corrections, too, which are important, but we could not find one, or a few parameters, the changes of which would reproduce tendencies of electrical transport properties.

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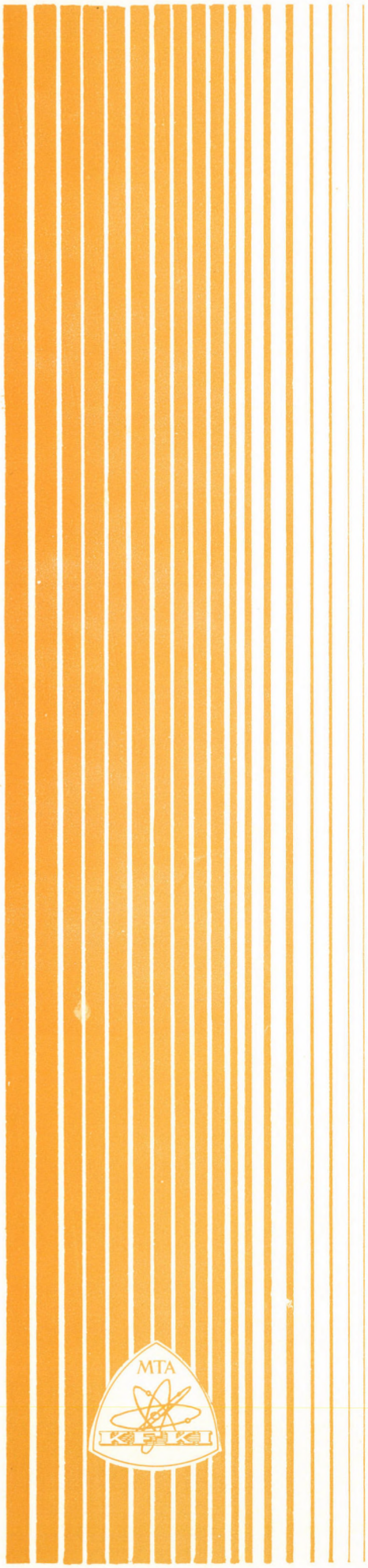

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