Influence of local and quasilocal impurity vibrational states on thermodynamic and superconducting properties of metallic glasses

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The specific heat of $Mg_{70}Zn_{30}$, $(Mg_{70}Zn_{30})_{95}Pb_5$, $Zr_{75}Rh_{25}$, and $(Zr_{75}Rh_{25})_{93}Be_7$ alloys was determined at temperatures 2–700 K in the amorphous and metastable crystalline states. The vibrational specific heat of ternary alloys $(Mg_{70}Zn_{30})_{95}Pb_5$ and $(Zr_{75}Rh_{25})_{93}Be_7$ exhibited singularities both in the amorphous and metastable crystalline states and these were due to specific quasilocal and local vibrations of the impurity ions relative to Pb and Be, respectively. A comparative analysis was made of the perturbing influence of heavy and light impurity atoms in the amorphous and metastable crystalline phases on the temperature dependence of the specific heat and on the superconducting transition temperature.

I. INTRODUCTION

Metals containing impurities, particularly substitutional impurities, are the simplest disordered metallic systems. It is known that introduction of even a small amount of an impurity with a very different mass into a metal affects the low-temperature behavior, altering significantly the thermodynamic, transport, and superconducting properties. A local change in the electron subsystem of a metal occurs near an impurity and this alters the electron spectrum as a whole. In the case of a phonon spectrum there are usually specific vibrational states associated with impurities. A light impurity in a "heavy lattice" gives rise to local vibrations beyond the edge of the allowed spectrum of the initial crystal, whereas a heavy impurity in a "light lattice" gives rise to resonance or quasilocal vibrations inside the allowed spectrum. The phonon spectrum as a whole also changes. This set of changes in the spectra of the excitation of a metal with an impurity alters the physical properties. Investigations of the influence of light and heavy impurities on the properties of regular (including metal) crystals have been going on for a long time. There have been many reviews of such work and the citations of these reviews can be found, for example, in Refs. 1–3. We shall mention simply that disorder in the distribution of impurities in substitutional systems excludes the possibility of appearance of a superstructure and a corresponding change in the spectra.

In contrast to crystalline systems containing impurities, amorphous metals (including metallic glasses), are an example of manifestation of extreme disorder because of the lack of the crystal structure. In the case of a metal or an alloy the transition to an amorphous state is accompanied (even without a change in the chemical composition) by a radical modification of the spectra of vibrational and electronic excitations, and corresponding changes in the thermodynamic, transport, and superconducting properties. In this case the primary reason for these changes is the disorder itself, giving rise to fluctuations of structure parameters and of corresponding parameters of the force interaction in an amorphous metal.

In view of this situation it is quite valid to ask the question: What is the influence of impurities with very different masses on the excitation spectra of an amorphous metal and, consequently, on its properties? Is it similar to the influence exerted on the properties of regular metals? Obviously, it is necessary to consider changes in all the thermodynamic, transport, and superconducting properties of an amorphous metal with an impurity in comparison with a pure amorphous metal and its crystalline analog. Studies of the influence of impurities with very different masses on the properties of an amorphous metal can shed additional light on the laws governing the formation of the impurity states in a metal and on the nature of the amorphous state of a metal itself.

The present paper reports the first stage of an investigation of the influence of impurity vibrational states on the properties of metallic glasses involving a calorimetric study of the manifestation of quasilocal vibrations in the specific heat of metallic glasses or their crystalline analogs. The initial systems were metallic glasses $Mg_{70}Zn_{30}$ and $Zr_{75}Rh_{25}$, investigated in detail earlier.[4,5] In the first of them the impurity was Pb and in the second it was Be. We selected these systems because of the large difference between the atomic masses of the impurity and the host system, and because these systems exhibit metastable crystalline phases of the same composition and with the same-range order.

II. PROPERTIES OF SAMPLES AND MEASUREMENT METHODS

The investigated systems were synthesized from metals of high initial purity: 99.99 at.% Zn, special purity (SOOO)Pb; vacuum distilled Mg; 99.98 at.% Zr; 99.95 at.% Rh; 99.5 at.% Be.

Amorphization of these alloys was induced in the usual way by rapid quenching from the liquid state on the outer surface of a rotating copper disk in an inert atmosphere. The samples were transformed from the amorphous to the metastable crystalline state by isothermal annealing. The composition was determined by chemical analysis and the structure by x-ray and electron diffraction methods.

The general form of the x-ray diffraction curves was typical of amorphous metals and showed no long-range or-

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TABLE I. Experimental data obtained by x-ray structure and differential thermal analyses, and densities of investigated systems.

System	2θ, K	$\Delta(2\theta)$, K	T_s, K	T_m, K	T_{ann}, K	d_a, g/cm^3	d_m, g/cm^3
$Zr_{75}Rh_{25}$	37	5	~1263	726	803	7,52	7,64
$(Zr_{75}Rh_{25})_{93}Be_7$	46.5	6.5	~1270	722	798	6,20	6,26
$Mg_{70}Zn_{30}$	38	5.6	463	376	433	2.98	3,05
$(Mg_{70}Zn_{30})_{95}Pb_5$	37.5	6	473	~372	443	3.56	3.24

der. The positions of the first wide maximum (2θ) and the widths of the maxima at half-amplitude $\Delta(2\theta)$ were determined (Table I). Identification of the metastable crystalline phases was made on the basis of an x-ray structure analysis which confirmed the published data on these systems. The structure of the samples in the metastable crystalline state was the same as those of $Mg_{51}Zn_{20}$ (Ref. 6) in the case of the $Mg_{70}Zn_{30}$ system, whereas in the case of the $Zr_{75}Rh_{25}$ system the structure was of the $E9_3$ type.[7]

The annealing temperatures T_{ann} and the temperatures of the transitions to the metastable state T_m and to the stable crystalline phase T_s were determined by the method of differential calorimetry[8,9] and from the temperature dependence of the electrical resistivity (Table I).

The densities of the investigated samples in the amorphous d_a and metastable crystalline d_m phases were found by weighing in air and in CCl_4. There was a reduction in the densities of both systems on transition to the amorphous state. The exception to this rule was the $(Mg_{70}Zn_{30})_{95}Pb_5$ system.

The temperature dependence of the specific heat of the samples was determined by the classical adiabatic method with pulsed heating in the interval 2–100 K in the absence of a magnetic field and in a magnetic field 6 T in the temperature range 2–7 K. The random error in the determination of the specific heat was 1–2% in the range 2–5 K and 0.4–1% in the range 5–100 K. Measurements of the specific heat in the 100–700 K range were made by the method of differential calorimetry.[10] which could be applied to samples of mass 10–200 mg and was subject to an error of $\approx 1.5\%$. More detailed descriptions of the methods used in the preparation of the samples and in the subsequent measurements were given in Refs. 11 and 12.

III. RESULTS OF MEASUREMENTS AND DISCUSSION

The temperature dependences of the specific heat of the original $Mg_{70}Zn_{30}$ and $Zr_{75}Rh_{25}$ samples and of $(Mg_{70}Zn_{30})_{95}Pb_5$ and $(Zr_{75}Rh_{25})_{93}Be_7$ with impurities were determined in the amorphous and metastable crystalline states at temperatures 2–20 K (they are plotted as dependences of C_p/T on T^2 in Fig. 1). We also found (Table II) the characteristic temperatures $\Theta(0)$ at low temperatures and Θ_{ht} at high temperatures, the critical temperatures of the transition to the superconducting state T_c, and the electron specific heat coefficient $\gamma(0)$ and γ_{ht}. As expected (Fig. 1), introduction of a light impurity (Be) into the $Zr_{75}Rh_{25}$ system in the amorphous and metastable crystalline states reduced the low-temperature specific heat of the system with an impurity, compared with the initial system. The scale of this change was approximately equal to the impurity concentration. On the other hand, introduction of a heavy impurity (Pb) into the system $Mg_{70}Zn_{30}$ increased the low-temperature specific heat of the system with an impurity in both (amorphous and metastable crystalline) states, compared with the initial samples. The scale of this change was considerably greater than the impurity concentration.

We shall now consider the changes in greater detail applying the traditional (in the analysis of the specific heat of systems with impurities) temperature dependence of the relative change in the vibrational specific heat normalized to the impurity concentration:

$$\frac{\Delta C_v(T)}{C_v(T)}=\frac{C_v(T)-C_v^{(0)}(T)}{C_v^{(0)}(T)\eta},$$

where $C_v(T)$ is the vibrational specific heat of a system with an impurity; $C_v^0(T)$ is the vibrational specific heat of the host system; η is the impurity concentration.

In the case of the $Zr_{75}Rh_{25}$ and $(Zr_{75}Rh_{25})_{93}Be_7$ systems with a known superconducting transition temperature the vibrational component was determined by subtracting the electron contribution from the total specific heat and allowing for the influence of the temperature-dependent electron-ion interaction. The method used in the calculation of the temperature dependence of the electron specific heat of the superconducting alloys affected by the electron-ion interaction was based on the theory of Ref. 13 and was described in Ref. 14. In the case of $Mg_{70}Zn_{30}$ and $(Mg_{70}Zn_{30})_{95}Pb_5$ systems, characterized by small values of the electron specific heat and a weak electron-phonon interaction, separation of the vibrational component was carried out in a traditional manner, i.e., without allowance for the temperature dependence of $\gamma(0)$:

$$C_v(T)=C_p(T)-\gamma(0)T$$

[$C_p(T)$ are the experimental values of the specific heat]. The results of such an analysis of the experimental data are presented in Fig. 2.

We shall begin by considering the influence of a light substitutional impurity on the properties of the $Zr_{75}Rh_{25}$ system, namely the temperature dependence of the specific heat and the temperature of the transition to the superconducting state in the metastable crystalline phase of $Zr_{75}Rh_{25}$ with the $E9_3$ structure.[15] The temperature dependence of the relative change in the vibrational specific heat is in this case represented by curve 4 in Fig. 2. Introduction of the light impurity Be into the metastable crystalline phase of $Zr_{75}Rh_{25}$ produces a negative relative change in the low-temperature vibrational specific heat with a minimum at ≈ 8 K. The changes in $\Theta(0)$ and Θ_{ht} indicate that the vibration-

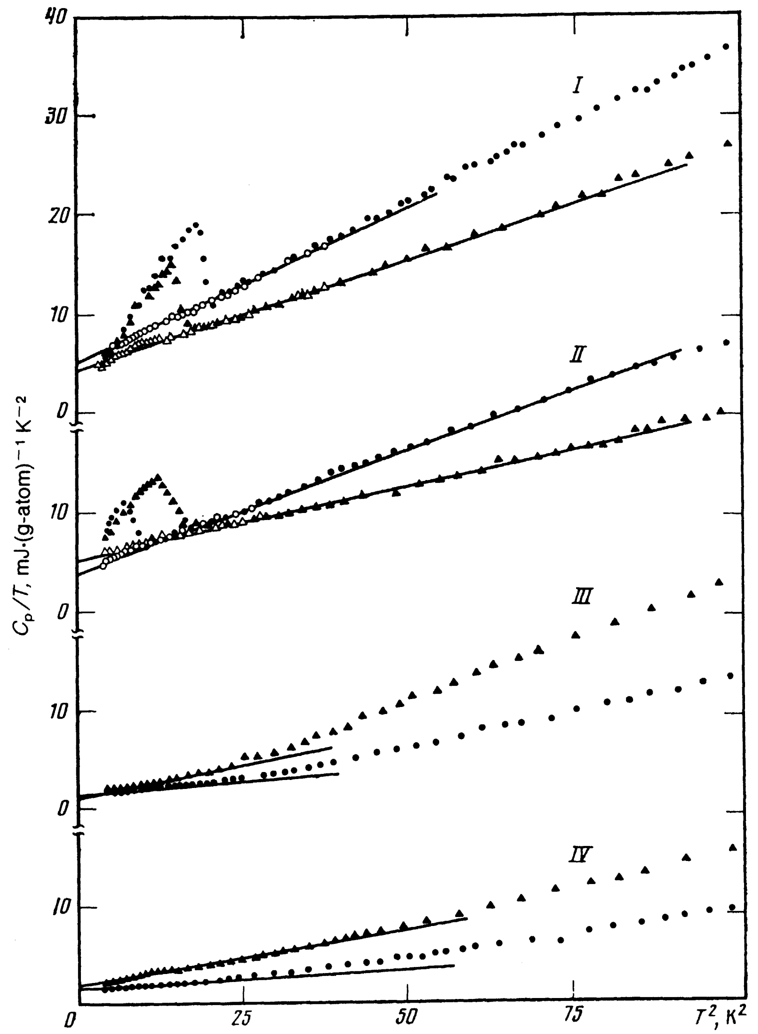

FIG. 1. Temperature dependences of the specific heat of the alloys $Zr_{75}Rh_{25}$ and $(Zr_{75}Rh_{25})_{93}Be$: I) amorphous states; II) metastable crystalline states; $Mg_{70}Zn_{30}$ and $(Mg_{70}Zn_{30})_{95}Pb_5$: III) amorphous state; IV) metastable crystalline state: ●) initial system; ▲) system with an impurity. Open symbols represent measurements in a magnetic field of 6 T.

al spectrum of the system with an impurity is harder than that of the initial impurity-free metastable crystalline phase. This result corresponds, in spite of the weak sensitivity of the low-temperature specific heat and the influence of light impurities, to the hypothesis that a local vibration region appears beyond the limit of the allowed spectrum of the metastable crystalline phase with an impurity. Introduction of the impurity is accompanied by an increase in the density of the electron states on the Fermi surface, which should result in strong softening of the vibrational spectrum of the system with an impurity and should increase its low-temperature specific heat. However, since this does not occur, it follows that the general change in the vibrational spectrum is dominated by the appearance of a local vibration, as illustrated clearly by an increase in $\Theta_{\rm ht}$ which can be interpreted as a shift of the high-frequency part of the vibration spectrum (since $\Theta_{\rm ht}^2 \propto \langle \omega^2 \rangle$) to the range of higher energies.

Calculations carried out on the basis of the above model describe satisfactorily the experimental dependence of the relative change in the vibrational specific heat (curve 4' in Fig. 2).

On the whole, in accordance with the predictions of Ref. 2, T_c of the system with the impurity increases by 30%

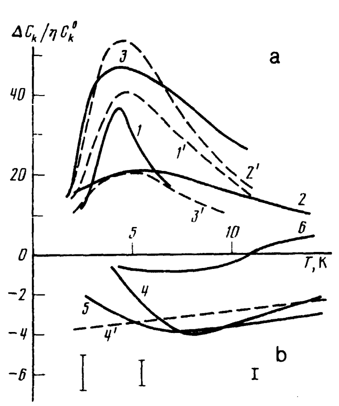

FIG. 2. Temperature dependences of the relative change in the vibrational specific heat of $Mg_{70}Zn_{30}$ (a) and $Zr_{75}Rh_{25}$ (b) alloys resulting from introduction of heavy and light impurity atoms Pb and Be, respectively, in the metastable crystalline (1,4) and amorphous (2,5) states. Curves 3 and 6 correspond to the case when the impurity system is amorphous and the initial system is metastable crystalline. The dashed curves represent the dependences calculated assuming the following values of the parameters: 1') $\Theta = 140$ K, $\gamma_1/\gamma_0 = 0.5$; 2') $\Theta = 140$ K, $\gamma_1/\gamma_0 = 0.4$; 3') $\Theta = 120$ K, $\gamma_1/\gamma_0 = 0.8$; 4') $\Theta = 204$ K, $\gamma_1/\gamma_0 = 1.6$. The confidence interval in the determination of $\Delta C_v(T)/\eta C_v^0(T)$ at several temperatures is shown below.

TABLE II. Parameters determined from low- and high-temperature specific heats.

System	State	T_c, K	$\Theta(0)$, K	$\gamma(0)$, mJ ·(g-atom)$^{-1}$ ·K^{-2}	Θ_{ht}, K	γ_{ht}, mJ ·(g-atom)$^{-1}$ ·K^{-2}
$Zr_{75}Rh_{25}$	AM	4.23	190	5.10	289	3.1
	MC	2.92	204	4,00	277	2,5
$(Zr_{75}Rh_{25})_{93}Be_7$	AM	3.96	204	4.40	306	2.7
	MC	3.83	234	5,30	~320	3.2
$Mg_{70}Zn_{30}$	AM	0.11 *	305	1.32	313	1,0
	MC	–	356	1,52	323	–
$(Mg_{70}Zn_{30})_{95}Pb_5$	AM	–	240	1,18	308	–
	MC	–	253	1,78	326	–

*The value of T_c were taken from Ref. 20 and the state in second column is either amorphous (AM) or metastable crystalline (MC).

because of an increase in the electron density of states, in spite of hardening of the vibrational spectrum. The nature of the interaction in these systems is intermediate because $\Delta C/\gamma T_c$ exceeds 1.43.

Introduction of the light impurity Be into the amorphous phase $Zr_{75}Rh_{25}$ produces practically the same results (curve 5 in Fig. 2 and Table II) in respect of the low-temperature specific heat. However, a minimum in the relative change of the vibrational specific heat is broader and the general hardening of the spectrum is less marked, as judged by a comparison of the values of $\Theta(0)$ and Θ_{ht} for the impurity and initial amorphous $Zr_{75}Rh_{25}$ phases. This happens under the conditions when the introduction of the light impurity into the $Zr_{75}Rh_{25}$ amorphous phase is accompanied by a fall of the density of the electron states on the Fermi surface, which tends to harden the vibrational spectrum because of the weaker screening of the ion-ion interaction. Nevertheless, in the case of the amorphous phase the local vibrations of the light impurities alter less the vibrational spectrum primarily because the conditions of formation of local vibrational states are more difficult to satisfy in the amorphous phase.

Figure 3 shows smoothed-out approximate functions representing the energy density of the vibrational states in the metallic glasses $Zr_{75}Rh_{25}$ and $(Zr_{75}Rh_{25})_{93}Be_7$, reconstructed from their specific heat measured in a wide range of temperatures by a method similar to that described in Ref. 14. The inset in Fig. 3 shows the net change in the density of the vibrational states in $Zr_{75}Rh_{25}$ due to the introduction of the Be atoms. We can see that the Be atoms give rise to local vibrations in the high-frequency part of the spectrum and these are separated by a gap from the range of vibrations of the host atoms; moreover, the density of the low-frequency vibrational states is reduced.

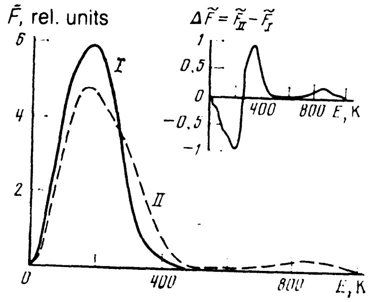

FIG. 3. Approxiamte functions describing the density of the vibrational states $\tilde{F}(\omega)$ in amorphous $Zr_{75}Rh_{25}$ (continuous curve) and $(Zr_{75}Rh_{25})_{93}Be_7$ (dashed curve) alloys. The inset is the difference between the functions $F(\omega)$ for $Zr_{75}Rh_{25}$ and $(Zr_{75}Rh_{25})_{93}Be_7$.

In contrast to the metastable crystalline phase of $Zr_{75}Rh_{25}$, introduction of the light impurity Be into the amorphous phase reduces T_c. This is facilitated by the hardening of the vibrational spectrum and by a reduction in the density of the electron states on the Fermi surface.

If we analyze the data on the amorphous $Zr_{75}Rh_{25}$ system with the impurity, but start with the impurity-free metastable crystalline phase, i.e., if we consider the simultaneous influence of the amorphization and of the introduction of the light impurity, we can see that these two processes practically compensate one another. Hardening of the vibrational spectrum which accompanies introduction of the light impurity is practically negligible against the background of the softening of the low-energy part of the spectrum as a result of the amorphization. We can see from curve 6 in Fig. 2 that the relative change in the low-temperature vibrational specific heat is slight, its sign is variable, and it depends weakly on temperature. The low-temperature value of $\Theta(0)$ is practically unaffected. A considerable rise of T_c in the case of the metastable crystalline $Zr_{75}Rh_{25}$ system on introduction of the Be impurity and on amorphization is primarily due to an increase in the density of the electron states.

It is easier to analyze the influence of a heavy impurity on the thermodynamic properties of the system than the influence of a light impurity, because introduction of a heavy impurity creates quasilocal vibrations which are manifested most clearly by a change in the low-temperature specific heat. In fact, when temperature is increased, the contribution to the specific heat made by an impurity atom and its environment vibrating at a frequency close to quasilocal vibrations increases and the value of $\Delta C_v(T)/\eta C_v^0(T)$ passes through a maximum, since the impurity contribution rapidly reaches the classical limit and the specific heat of the initial system continues to rise. The position and width of the maximum of the relative change in the vibrational specific heat make it possible to draw reliable conclusions on the characteristics of quasilocal vibrations.

We shall describe the experimental data by confining ourselves to a qualitative analysis of the influence of an impurity, the atomic weight of which differs considerably from the atomic weight of the host atoms and we shall use a model[16] of the Debye spectrum of vibrations and of the interaction of this impurity with the nearest neighbors. In this model the vibrational spectrum is approximated by the expression

$$g(\omega)\simeq\frac{3\omega^2(1-\eta)}{\omega_g{}^3}+\eta\frac{\Gamma}{(\omega-\omega^*)^2+\Gamma^2},\qquad(1)$$

where $\Gamma\sim\gamma_1^2/M$ is the width of a quasilocal vibration level; $(\omega^*)^2=\gamma_1/M$ is the position of a quasilocal vibration level; M is the mass of an impurity atom; γ_1 is the force constant of the interaction between the impurity and the host matrix; γ_0 is the force constant of the interaction between the host matrix atoms themselves; the impurity-induced change in the vibrational specific heat is

$$\frac{\Delta C_v(T)}{C_v{}^0(T)}\sim 3\eta\frac{k_B}{T}\int\limits_0^\infty d\omega^2\,\omega^2\,\Delta g(\omega)\,\frac{\exp(\omega/T)}{(\exp(\omega/T)-1)^2},$$

where $\Delta g(\omega)$ corresponds to the first term in Eq. (1). In this system there are two fitting parameters: the average Debye frequency $\tilde{\omega}$ and the force constant γ_1. We shall analyze the influence of the heavy impurity Pb on the properties of the $Mg_{70}Zn_{30}$ system on the basis of the above model.

Introduction of the heavy Pb impurity into the $Mg_{70}Zn_{30}$ system in the metastable crystalline phase results in the expected appearance of a quasilocal vibration (curve 1 in Fig. 2 and Table II) in the form of a maximum of the temperature dependence $\Delta C_v(T)/\eta C_v^0(T)$ and it causes general softening of the vibrational spectrum both because of the appearance of quasilocal vibrations and also because of an increase in the density of the electron states on the Fermi surface. However, the width of the maximum is considerably less and the maximum itself is located at a lower temperature than in the case of quasilocal vibrations in the doped crystalline system.[17,18]

In accordance with the adopted model, the most satisfactory description of the temperature dependence of $\Delta C_v(T)/\eta C_v^0(T)$ is obtained for the average value of the Debye temperature ≈ 140 K in the case weakening by a factor of two of the local force constants for the interaction of the impurity with the host matrix: $\gamma_1/\gamma_0\approx 0.5$. This means that the functions describing the density of the vibrational states have a singularity corresponding to quasilocal vibrations which is anomalously narrow, because it is proportional to γ_1^1/M and, consequently, a Pb impurity atom is located in the metastable crystalline phase of $Mg_{70}Zn_{30}$ at low-symmetry positions of the structure, resembling the situation in the case of weakly bound Pb atoms in Cevrel phases.[19]

In the case of the amorphous $Mg_{70}Zn_{30}$ phase (curve 2 in Fig. 2), introduction of such heavy Pb impurity atoms is accompanied by a weaker manifestation of quasilocal vibrations. A maximum of the temperature dependence of $\Delta C_v(T)/\eta C_v^0(T)$ is located at $T\approx 5.5$ K. It is broadened on the temperature scale and its amplitude is approximately half that for the metastable crystalline phase. The observed slight shift of the maximum toward higher temperatures is in qualitative agreement with the reduction in the density of the electron states in the amorphous system with an impurity, compared with an impurity-free amorphous system.

A comparison of the experimental curve 2 with the theoretical dependence calculated allowing for the renormalization of the vibration spectrum ($\Theta\approx 120$ K) shows that Pb is bound much more strongly in an amorphous system ($\gamma_1/\gamma_0\approx 0.8$) than in the metastable crystalline phase, for which we have $\gamma_1/\gamma_0\approx 0.5$. On the whole, a comparison with the experimental data on the metastable crystalline and amorphous phases shows that introduction of a heavy impurity into a metastable crystalline phase alters the average $\tilde{\Theta}(0)$ and local γ_1/γ_0 parameters much more than in the case of an amorphous system.

Curve 3 in Fig. 2 represents the combined effect of the influence of a heavy impurity and of the amorphization on the relative change in the vibrational specific heat in the $Mg_{70}Zn_{30}$ system. The observed large change in $\Delta C_v(T)/\eta C_v^0(T)$ is related to the introduction of the heavy impurity and the amorphization, in spite of a reduction in the density of the electron states on the Fermi surface.

Unfortunately, the absence of the data on T_c of the $(Mg_{70}Zn_{30})_{95}Pb_5$ system in the amorphous and metastable crystalline states prevented us from an analysis of the changes in T_c in the $Mg_{70}Zn_{30}$ system on introduction of the heavy Pb impurity atoms. In these cases the value of T_c was not observed right down to 0.4 K when measurements were made by the electrical resistivity and inductive methods.

CONCLUSIONS

The results of a comparative analysis of the experimental data and model calculations on the influence of light and heavy substitutional impurities on the temperature dependence of the specific heat or binary alloys in metastable crystalline and amorphous states allows us to draw the following conclusions.

1. Introduction into amorphous alloys of impurities with very different mass from the mass of the atoms in the host matrix alters, as in the case of crystalline systems, the temperature dependence of the specific heat in a way which is characteristic of light impurities (local vibrational states) and heavy impurities (quasilocal vibrational states). However, the manifestation of the perturbing influence of such impurities on the spectrum of the thermal vibrations and on the specific heat of amorphous systems is greatly weakened by fluctuations of the structure parameters and of the parameters of the force interaction itself. The changes occurring in the electron system enhance softening of the vibrational spectrum in the metastable crystalline state and compensate such softening in the amorphous state.

2. A change in the temperature of the transition to the superconducting state associated with the introduction of the light Be impurity atoms into the $Zr_{75}Rh_{25}$ alloy is dominated by the associated change in the electron subsystem. The change in the vibrational spectrum has less influence on the change in T_c.

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