

Low-frequency dynamics in metal alloys: “ferroelectric” anomalies at finite frequencies

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A giant absorption of the power of an electromagnetic field has been observed in metal alloys. The effect, resonant in the frequency, is observed in a narrow concentration interval near an electronic phase transition. The interrelation between the observed effect and a dipole-active low-frequency lattice dynamics is discussed.

Anomalies in the structural state of metals and alloys which are manifested as distinctive features in the quasielastic scattering of neutrons and Mössbauer radiation have recently attracted considerable interest.¹⁻⁴ A basic question is whether these anomalies result from static distortions or low-frequency dynamic processes (“low” here means in relation to the frequencies of phonons). These particular experimental techniques are not capable of resolving this question unambiguously, and they are especially unsuitable for studying the nature of the low-frequency dynamics, if such exists. The existence of a low-frequency dynamics might be manifested in a resonant interaction with an external electromagnetic field. In metallic systems, however, it would appear that it would be impossible to observe resonances of this sort because of the small skin thickness.

In this letter we are reporting the observation of a resonant absorption of the power of an electromagnetic field in the alloy systems $Ti_{1-x}Me_x$ (Me: V, Cr, Fe, Co, Mo) and $Zr_{1-x}Fe_x$ in narrow concentration intervals. The results show that the resonant interaction of the electromagnetic field with the low-frequency dynamics is observable because of a proximity to the electronic phase transition observed in Ref. 5. We reported similar results for conducting ceramics in Refs. 6 and 7. The giant absorption of electromagnetic power in systems with a metallic conductivity is thus an extremely common effect. This circumstance would seem to rule out a relationship between the dielectric anomalies in ceramic conductors^{6,7} and particular features of these materials (in particular, their granularity).

The effective conductivity $\sigma(\nu)$ was measured over the frequency range 0.7 GHz $\leq \nu \leq 11$ GHz, as in Ref. 6, by measuring the absorbed power by a method involving a combined transmission-reflection resonator⁷ at room temperature. To eliminate instrumental errors, we used a comparison method with several standards: Ag ($\rho = 1.35 \mu\Omega \cdot \text{cm}$), V ($16.8 \mu\Omega \cdot \text{cm}$), Ti ($43 \mu\Omega \cdot \text{cm}$), La ($58 \mu\Omega \cdot \text{cm}$), and Ge-As ($0.1 \Omega \cdot \text{cm}$ and $1 \Omega \cdot \text{cm}$). The standards were of the same shape and dimensions as the samples of the alloys being studied. For all of the standards, the conductivity values at all of the frequencies studied agreed within 5% with the static conductivity $\sigma(0)$.

Figure 1 shows the concentration dependence of the effective resistivity $\rho = 1/\sigma$ measured at a frequency of 6.2 GHz and also the dc resistivity for alloys of Ti with Fe and Mo and for Zr-Fe (similar results were found for the Ti-Cr and Ti-Co systems). In all the systems studied, there was a narrow concentration interval, in which $\rho(\nu)$ was two to four orders of magnitude higher than $\rho(0)$. The maximum of the ratio $\rho(\nu, X)/\rho(0, X)$ is reached at concentrations at which we observe the maximum of the residual resistivity and at which the temperature coefficient of the resistance goes

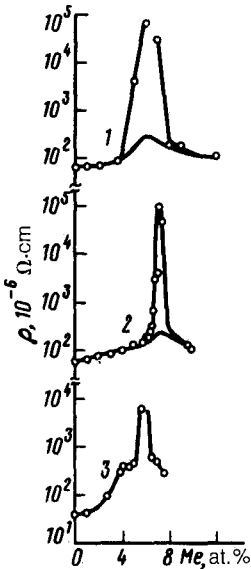


FIG. 1. Points: concentration dependence of the effective resistivity at the frequency 6.2 GHz. Solid lines without points: direct-current resistivity. 1—Alloys of Ti with Fe; 2—with Mo; 3— $Zr_{1-x}Fe_x$.

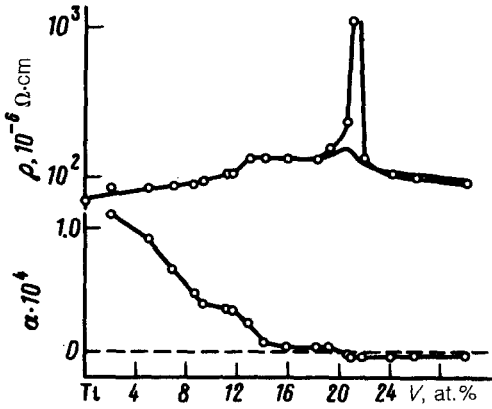


FIG. 2. Points—concentration dependence of the effective resistivity at the frequency 6.2 GHz; solid lines without points—dc resistivity; lower curve—temperature coefficient of the resistivity of $Ti_{1-x}V_x$ alloys.

negative. The situation is illustrated by Fig. 2, which shows, along with the values of $\rho(\nu)$ and $\rho(0)$ for the Ti-V system, the concentration dependence of the temperature coefficient of the resistance at room temperature, measured for the same samples at a direct current. According to Ref. 5, the concentration at which the sign of the temperature coefficient of the resistance changes in titanium alloys is a point of an electronic phase transition.

It can be seen from Fig. 3 that the frequency dependence $\sigma(\nu)$ is of a resonant nature. The resonant frequencies ν_0 for the various alloys differ slightly and are more than an order of magnitude greater than ν_0 for conducting ceramics.^{6,7} Consequently, we are observing a giant absorption of the power of a high-frequency field, of a resonant nature, near an electronic phase transition in these metal-alloy systems.

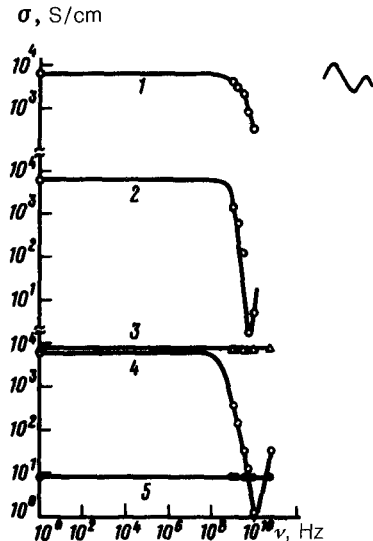


FIG. 3. Frequency dependence of the effective conductivity ($\sigma \equiv 1/\rho$) of the following alloys: 1—Ti-21 at.% V (the solid line without points shows the results of optical measurements in the IR range, reproduced from the data of Ref. 11); 2—Ti-6 at.% Fe; 3—Ti-10 at.% Mo; 4—Ti-7.4 at.% Mo; 5—Ge-As.

Since none of the alloys studied are ferromagnetic, the experimental results should be interpreted, as in Ref. 6, as the observation of an electromagnetic loss due to dipole moments which arise at a finite frequency,⁶ i.e., "ferroelectric" anomalies at finite frequencies in metal alloys.

A phenomenological model describing the observed effect was proposed in Refs. 6 and 7. It follows from the experimental results reported here that the function $\beta(\omega)$ in Eq. (3) of Ref. 6 is a resonant function of the frequency [β must be replaced by $-\beta$ in Eq. (2) in Ref. 6]. According to this model, the resonant electromagnetic loss in a metal stems from a combination of three factors: the existence of a low-frequency dynamics, a short mean free path of conduction electrons, $l \sim a$ (a is the lattice constant), and comparatively large values of the Debye screening length, $\lambda \gg a$. The conducting ceramics which were studied in Refs. 6 and 7 have yet to be studied thoroughly, and there is no independent experimental evidence for the occurrence of these factors, aside from the condition $l \sim a$, in those materials. The alloys discussed in the present letter, on the other hand, have been studied in vastly more detail, and there are grounds for believing that these factors are operating in these materials:

1. The results of Refs. 2, 3, and 8 point to the existence of a low-frequency dynamics for Zr alloys. The data of the present study should be regarded as direct experimental proof of the existence of a low-frequency dynamics in transition-metal alloys based on titanium and zirconium. The resonant frequency dependence of the absorption indicates that the low-frequency dynamics should be of an oscillatory rather than relaxation nature.

2. We know quite well that the condition $l \sim a$ holds for all of the alloys studied near the critical concentration for the electronic phase transition,⁹ X_c .

3. The Debye screening length is $\lambda \gg a$. For titanium- and zirconium-based alloys, this conclusion follows from the fact that an electronic phase transition occurs in these materials.⁵ The model of Refs. 5 and 9 for the electronic phase transition is assumed to have two electronic subsystems. In one of these subsystems an insulator-metal transition occurs at $X = X_c$, while the conductivity remains finite in the other subsystem. The static dielectric constant of the first subsystem becomes infinite, $\epsilon_0 \rightarrow \infty$, and the state density at the Fermi level, $N(E_F)$, decreases. The net result is an increase in $\lambda = [\epsilon_0/4\pi N(E_F)e^2]^{1/2}$. The structural feature in ϵ , according to the general analysis of Ref. 10, leads to the anomalies which are observed in the electronic and lattice properties of these alloys near^{9,5} X_c . The X dependence of $\rho(\nu)$ in Figs. 1 and 2 confirms the conclusions of the model of Refs. 5 and 9, since conditions (1) and (2) hold over a concentration region^{2,3,5} which is much wider than that in which the giant absorption is observed. The latter region is thus distinguished by specifically the condition that λ increases. In the course of the experiments, we determined the critical concentrations for the electronic phase transitions in the Ti-Mo and Zr-Fe alloys (Fig. 1).

With regard to the microscopic nature of the low-frequency dynamics, we see two possibilities. According to Ref. 8, the low-frequency dynamics might be a consequence of collective effects in the phonon subsystem and would be manifested by the appearance of some new time scales (for processes which are slow in comparison with

phonon times) and associated length scales (for regions which are large in comparison with a). In these regions the symmetry of the lattice is disrupted by dynamic processes, with the possible result that a polarization vector appears at a finite frequency in structures with an inversion center. One of the specific models for a low-frequency dynamics which was examined in Ref. 8—the model of a nonlinear phonon resonance—predicts that as the temperature is lowered to values below the energy of the phonons involved in the resonance (the quantum regime), the integral absorption intensity decreases substantially, and the resonance becomes sharply peaked in the frequency. At liquid-helium temperature the effect should then vanish in these alloys [but possibly not in the ceramics of Refs. 6 and 7, where there might be low-frequency “librational” vibrations (for example), associated with a copper-oxygen octahedron].

The second possibility is related to the need to develop a quantum model for the low-frequency dynamics on the basis of an examination of effects of the nature of a cooperative atomic tunneling in comparatively large regions $R > l$ (R is the length scale of the regions). The latter is required for an effective interaction of conduction electrons with dipole moments through the local field in the medium.^{6,7}

The model of Refs. 5 and 9 for the electronic phase transition seems to be suitable for a wider class of systems than simply titanium- and zirconium-based alloys, including both substitutional and implantation alloys. The result has been to stimulate a study of $\sigma(\nu)$ in A15 systems and in substoichiometric nitrides of transition metals. We have observed anomalies in $\sigma(\nu)$ of the type described above in nitrides of titanium, zirconium, and niobium and also (although considerably more weakly) in Nb₃Al and V₃Ga. These results will be reported and discussed in detail in another paper.

In summary, a low-frequency dynamics manifested by the existence of a dipole-active mode with a frequency $\nu_0 \sim 10^8 - 10^{10}$ Hz is a rather common effect among metallic systems which undergo an electronic phase transition of the type in Ref. 5. The effect which has been observed might be turned into the most effective tool for studying the nature of the low-frequency dynamics.

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