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We present expressions for the change of sound velocity and sound absorption due to phase fluctuations in a lattice of Josephson-coupled metallic grains. An approximate evaluation predicts measurable effects unique to granular superconductors.

RESEARCH ON GRANULAR metals has grown continuously since some of their properties were first reviewed about ten years ago in a very stimulating article by Abeles et al. [1]. Typically, these granular structures consist of small metal particles ($< 100 \text{ Å}$) embedded in an amorphous insulator. A different type of system is given by random mixtures of metals and insulators. Both classes of heterogeneous media have in common that their metal—volume fraction p can be changed systematically. This opens the possibility (a) to view their physical properties through the framework of percolation theory, (b) to see in them a realization of localization phenomena — or (c) to consider both aspects together [2].

Superconductivity in heterogeneous media is certainly of great interest. There is not yet a microscopic description for the full temperature range based upon the modified electron—phonon interaction. However, at low temperatures one often studies a model granular superconductor which is obtained by integrating away all microscopic degrees of freedom thereby arriving at an effective energy

$$H_{\text{Phase}} = H_{\text{Coulomb}} + H_{\text{Josephson}}. \tag{1}$$

Here the individual grains are already superconducting and are coupled by their Josephson phases

$$H_{\text{Josephson}} = -\sum_{ij} E_{ij} \cos(\phi_i - \phi_j), \tag{2}$$

and

$$H_{\text{Coulomb}} = \frac{1}{2} \sum_{ij} B_{ij} p_i p_j, \quad p_i = -i \frac{\partial}{\partial \phi_i}, \tag{3}$$

simulates the charging energy due to transfer of Cooper pairs with B being proportional to the inverse of the capacitance matrix of the collection of small particles [3].

The Hamiltonian of equation (1) contains very rich structures with respect to phase coherence, randomness,

dimensionality and magnetic field dependence [4]. In this short paper we want to discuss still another aspect which is related to the elasticity of the granular metal.

Obviously the electronic properties of every granular metal will depend on tunneling between grains and are therefore strongly influenced by variations of the intergrain separations. This observation suggests that pressure effects should exist — at least for hard grains embedded in a soft matrix. In particular near the threshold value of the Josephson coupling E a granular superconductor described by equation (1) should be pushed through the phase boundary [5] into the coherent state by applying pressure.

These remarks also lead immediately to a coupling between Josephson phases and sound, since the effect of a sound wave is to modulate the interparticle spacings in the array of superconducting grains. We can assume that the Josephson coupling varies exponentially with distance r_{ij} of neighbouring grains $E_{ij} \sim \exp(-\kappa r_{ij})$. The position r_i of grain i becomes $r_i + u_i$ when a sound wave of wave vector q and frequency ω travels through the system. For simplicity we will regard H_{Phase} of equation (1) as a periodic model of identical grains with spacing d. This gives for $qd \ll 1$ an interaction

$$H_{\text{int}} = E\kappa \sum_{\langle ij \rangle} (u_i - u_j) \cdot e_{ij} \cos(\phi_i - \phi_j), \tag{4}$$

where the sum is over nearest neighbours and e_{ij} is the unit vector in direction $r_i - r_j$.

The surrounding medium will mediate an elastic interaction between grains of mass M which is expressed by the harmonic Hamiltonian

$$H_{\text{elast}} = \sum_i \frac{p_i^2}{2M} + \frac{1}{2} \sum_{ij} u_i \underline{C} u_j. \tag{5}$$

The corresponding long-wavelength phonons are identified with ultrasonic waves, and the total Hamiltonian $H = H_{\text{Phase}} + H_{\text{elast}} + H_{\text{int}}$ is then a simple model for the coupling of ultrasound to the phase degrees of freedom.

We should mention that a similar model was considered a long time ago for compressible spin systems where the coupling originates from the distance dependence of the exchange interaction [6].

Acoustic properties of interest are contained in the phonon propagator $D(i, t; j, t') = -i \langle T(u_i(t)u_j(t')) \rangle$. Perturbation theory with respect to H_{int} of equation (4) leads in a standard way to damping and frequency shift of phonons. For long wavelengths the frequency shift determines the change in sound velocity c via the linear relation $\Delta \omega = q \cdot \Delta c$. In this way we obtain for longitudinal waves the relative change in sound velocity [7]

$$\frac{\Delta c}{c} = \frac{(E\kappa d)^2}{\hbar Mc^2} \frac{1}{N} \sum_{\substack{\langle ij \rangle \\ \langle lm \rangle}} (e_{ij} \cdot \hat{q})^2 (e_{lm} \cdot \hat{q})^2$$

$$\exp[-i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_m)] \operatorname{Re} G_{lm}^{ij}(\omega) \qquad (6a)$$

and the sound absorption coefficient

$$\alpha = -q \frac{(E\kappa d)^2}{\hbar Mc^2} \frac{1}{N} \sum_{\substack{\langle ij \rangle \\ \langle lm \rangle}} (e_{ij} \cdot \hat{q})^2 (e_{lm} \cdot \hat{q})^2$$

$$\exp[-i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_m)] \operatorname{Im} G_{lm}^{ij}(\omega). \qquad (6b)$$

bulk susceptibility for small $\epsilon = (T - T_c)/T_c$ obtained by means of a time-dependent Ginzburg–Landau treatment [9]. $\Gamma_0^{-1} \sim \epsilon^{-1}$ is the relaxation time and $\xi \sim \epsilon^{-1/2}$ the correlation length. Note that we transfer here the bulk behaviour to the paracoherent region of the granular model of equation (1) where the BCS-order-parameter amplitude Δ is non-fluctuating whereas the phases fluctuate and drive the system into the coherent state as $\epsilon \rightarrow 0$ [10]. It is clear that we pass with this approximation from the original quantum mechanical behavior contained in G of equation (7) to a phenomenological description.

The expressions in equation (6) can now be explicitly calculated [7]:

$$\frac{\Delta c}{c} = -\frac{4}{\pi^2} \frac{k_B T}{Mc^2} \left(\frac{E\kappa d}{\Delta^2 N(0)}\right)^2 \frac{(qd)^3}{\Omega^2} I_2, \qquad (9a)$$

$$\alpha = q\Omega \left(\frac{-\Delta c}{c}\right) I_1/I_2, \qquad (9b)$$

where

$$I_n(\epsilon) = \int_0^{k_D/q} dx \int_{-1}^{1} d\mu \frac{x^2 [Q^2 + 2(\epsilon + Q^2 x^2 - Q^2 x\mu)]^n}{(\epsilon + Q^2 x^2)(\epsilon + Q^2 x^2 - 2Q^2 x\mu + Q^2)(1 + \Omega^{-2}[Q^2 + 2(\epsilon + Q^2 x^2 - Q^2 x\mu)]^2)}. \qquad (10)$$

Here N is the number of grains and $G(\omega)$ is the Fourier transform of the generalized phase-phase susceptibility

$$G_{lm}^{ij}(t) = -i\theta(t) \langle [\cos(\phi_i(t) - \phi_j(t)), \cos(\phi_l(0) - \phi_m(0))] \rangle, \qquad (7)$$

the thermal average being taken with respect to H_{Phase} of equation (1).

In previous work [8] the absorption coefficient α was obtained by a slightly different method, and it was argued qualitatively that in the critical regime approaching the transition temperature T_c from above α should increase as $(T - T_c)^{-y}$ with $y \sim 1$. Here we are mainly interested in the sound velocity which we want to calculate together with α for $T > T_c$ using the simplest approximations for phase fluctuations. This is done in two steps.

First, G of equation (7) is factorized in all possible ways using $\langle \cos \phi_i \rangle = 0$ in the paracoherent state $(T > T_c)$. As a consequence the order-parameter susceptibility $\chi(r_{ij}, t) = -i\theta(t) \langle [\cos \phi_i(t), \cos \phi_j(0)] \rangle$ becomes the essential quantity.

Second, the Fourier transform of χ is written in the simple form

$$\chi(\mathbf{q}, \omega) = [N(0)|\Delta|^2 (1 - i\omega/\Gamma_0 + \xi^2 q^2)]^{-1}. \qquad (8)$$

This ansatz is suggested by the result for the pairfield

Here k_D represents the Debye-wave vector of the grain lattice and $\Omega = \epsilon\omega/\Gamma_0$, $Q^2 = \epsilon(\xi q)^2$ are temperature independent, dimensionless quantities.

An analysis of equation (10) shows that Δc and α behave approximately as

$$-\Delta c \sim (T - T_c)^{-1/2}, \qquad \alpha \sim \omega^2 (T - T_c)^{-3/2}, \qquad (11)$$

for $10^{-4} \lesssim \epsilon \ll 1$ and approach finite values at T_c.

In order to obtain an estimate of the magnitude we use typical values for granular aluminum, $T_c \sim 2$ K, grain diameter $D \sim 30$ Å, distance between grain centers $d \sim 50$ Å. According to [11]: $\epsilon^{1/2}\xi = 0.85(\xi_0 l)^{1/2}$ with l of order $d/2$. With $\kappa d \sim 1$ and $\omega = 10$ MHz the numerical evaluation in equation (9) gives (see Fig. 1)

$$-\frac{\Delta c}{c} \gtrsim 10^{-4} \qquad \text{for} \qquad \epsilon < 10^{-3},$$

$$\alpha \gtrsim 10^{-3} \text{ cm}^{-1} \qquad \text{for} \qquad \epsilon < 10^{-4}. \qquad (12)$$

Within the uncertainty in the value of the reciprocal tunneling length κ, this theory indicates that at least Δc should be detectable in the critical regime.

Ultrasound experiments have been reported by Levy and coworkers who measured the attenuation of surface waves in granular metal films. No enhancement of α as indicated by equation (11) has been observed so far, and it appears that percolation arguments [12] might be
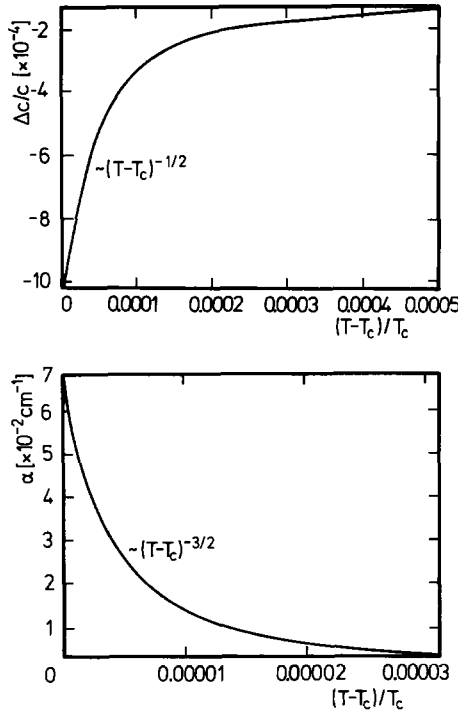
Fig. 1. Relative change in sound velocity and sound absorption coefficient due to interaction with Josephson phases in granular Al/Al_2O_3 (see [11]) near the transition temperature T_c.

more appropriate for such substrate-coupled films than our idealized grain-lattice model.

The sound velocity in a granular metal near the transition into the superconducting state has to our knowledge never been studied experimentally. According to equation (12) a measurable effect should exist which is not present at T_c in usual homogeneous superconductors. We are aware that the change in sound velocity which we predict follows from drastic approximations

on an idealized model. In spite of this we hope that our considerations will stimulate ultra sound velocity measurements on suitable granular probes.

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