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Probing porous media with superfluid acoustics

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Abstract. We discuss those properties of porous media which can be deduced from experiments using measurements of superfluid 1st, 2nd, 4th, and 3rd sound; we also explore the transferability of these results to other transport experiments, especially the acoustic properties of porous media saturated with Newtonian fluids. Many of the relevant geometrical parameters are those which arise in a canonical electrical conductivity problem in which the porous solid is insulating, the pore fluid is conducting, and there is an additional surface conductivity lining the walls of the pore space. The most important geometrical parameters are the three-dimensional tortuosity of the pore space, α_3, the two-dimensional tortuosity of the pore/grain interface, α_2, and Λ, which is a well-defined measure of dynamically connected pore sizes.

1. Introduction

Historically, the understanding of superfluid ^{4}He in terms of the macroscopic two-fluid equations of motion was established in no small measure by means of experiments in porous media and other restricted geometries. It is now possible to turn this situation around and use the superfluid as a probe of the transport properties of porous media. There are several parameters, characteristic of the pore geometry, which describe aspects of many of these transport phenomena. In order to introduce them, it is pedagogically useful to describe a paradigm problem in electrical conduction. For a more complete review of these effects, we refer the reader to an earlier article [1].

Consider, then, the problem of electrical conduction in a porous medium in which the solid phase is electrically insulating, the pore fluid has a conductivity σ_f and the walls of the pore space are coated with a surface conductor of strength Σ_s. In figure 1 we plot the results of calculations of the effective conductivity, σ_{eff}, on two model porous media of differing porosities, ϕ, wherein Σ_s is held constant whilst σ_f is varied (from [2]). The calculated conductivity σ_{eff} is a complicated function of σ_f and of Σ_s which simplifies in two limiting cases as follows.

(i) If conduction is dominated by the pore fluid, with the surface conductivity as a weak perturbation, we have

$$\sigma_{eff} = F^{-1}\left(\sigma_f + \frac{2\Sigma_s}{\Lambda}\right) + \sigma_f O\left(\frac{\Sigma_s}{\sigma_f \Lambda}\right)^2 \tag{1a}$$

where F and Λ are constants characteristic of the porous medium. In this limit conduction is nearly proportional to σ_f and the effects of Σ_s are a perturbation. In

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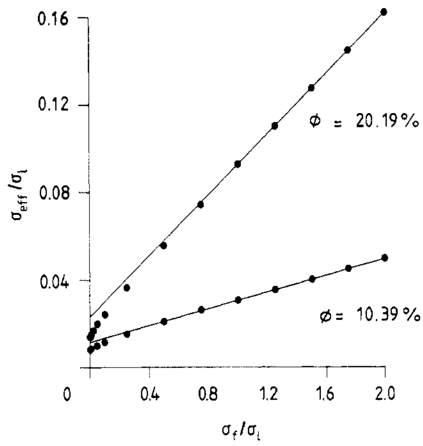

Figure 1. Calculated conductivity, σ_{eff}, as a function of pore-fluid conductivity, σ_f, in the grain consolidation model with a fixed value of surface conductivity, Σ_s. The data points are the results of numerical calculations and the straight lines are equation (1). After [2].

fact, it is an exact result [2, 3] that Λ can be related to the microscopic potential $\psi_0(\boldsymbol{r})$ which would exist in the *absence* of the surface mechanism

$$\frac{2}{\Lambda} = \int \mid \nabla\psi_0(\boldsymbol{r}) \mid^2 \, \mathrm{d}S \left(\int \mid \nabla\psi_0(\boldsymbol{r}) \mid^2 \, \mathrm{d}V_{\mathrm{p}} \right)^{-1} . \tag{1b}$$

Λ is a measure of the size of the dynamically connected pore space. It is defined in terms of a weighted surface-to-pore-volume ratio in which only those parts of the pore space which carry current are counted; in the special case of a pore space consisting of winding cylindrical tubes of radius R, then $\Lambda \equiv R$. It will prove convenient to define the three dimensional tortuosity of the pore space: $\alpha_3 \equiv F\phi$ where ϕ is the porosity. The straight lines implied by equations (1*a*) and (1*b*) are plotted in figure 1.

(ii) The opposite limit, in which conduction is dominated by the surface mechanism, is formally similar to that given above (see [1]), namely

$$\sigma_{\mathrm{eff}} = f^{-1}\left(\Sigma_{\mathrm{s}} + \frac{\lambda\sigma_{\mathrm{f}}}{2}\right) + \Sigma_{\mathrm{s}} \mathrm{O}\left(\frac{\lambda\sigma_{\mathrm{f}}}{\Sigma_{\mathrm{s}}}\right)^2 . \tag{2}$$

It will prove convenient to define the two-dimensional tortuosity of the pore surface: $\alpha_2 \equiv f(S/V)$ where S/V is the surface-to-total-volume ratio.

The reason for these arcane definitions is that α_3, Λ, and α_2 are relevant to a large class of experiments on porous media, notably acoustics and diffusion, and that the values deduced from one experiment are directly transferable to the others. In the rest of this article, we shall review those results.

2. Dynamic permeability

If a porous medium, saturated with a viscous fluid, is subjected to an oscillatory pressure gradient, then the induced fluid flow will also be oscillatory and proportional to the pressure gradient

$$\phi \boldsymbol{v} = -\frac{\tilde{k}(\omega)}{\eta} \nabla P \tag{3}$$

where η is the viscosity and $\tilde{k}(\omega)$ is the dynamic permeability; $\tilde{k}$ is frequency dependent in the region where the viscous skin depth is comparable to Λ. Indeed, the high-frequency limit of $\tilde{k}(\omega)$ is *exactly* related [3] to α_3 and to Λ. A model [3] for $\tilde{k}(\omega)$ based on a variety of exact results (notably the low-frequency and the high-frequency behaviour) is

$$\tilde{k}(\omega) = k_0 \left[\left(1 - \frac{4\mathrm{i}\alpha_3^2 k_0^2 \rho_\mathrm{f} \omega}{\eta \phi^2 \Lambda^2} \right)^{1/2} - \frac{\mathrm{i}\alpha_3 k_0 \rho_\mathrm{f} \omega}{\eta \phi} \right]^{-1} \tag{4}$$

where k_0 is the DC permeability. This function works well when compared against calculations based on a periodic array of truncated spheres or octahedra [4], and against experimental measurements taken on sphere packs [5] as shown in figure 2. (See [6].)

3. 1st and 2nd sound

Below a temperature $T_\lambda = 2.17$ K, ^{4}He undergoes a phase transition in which it behaves something like a miscible mixture of two fluids: a normal fluid having a small but finite viscosity, and a superfluid fraction which has exactly zero viscosity. Because there are two degrees of freedom there are two acoustic normal modes: 1st sound, the usual pressure/density wave and 2nd sound, in which the temperature and entropy propagate as waves. There are several advantages in using He II (as it is called) to probe porous media [1]:

(i) It is much more compressible than most solids, so the porous medium may be considered perfectly rigid.

(ii) The effects of viscosity can be varied simply by changing the temperature (i.e. by varying the normal-fluid/superfluid ratio).

(iii) For the same reason, the viscous skin depth, which is the yardstick for probing the porous media, can be varied as a function of temperature. As a point of information, the viscous skin depth of the normal fluid ranges from 0.1 μm to 10.0 μm for temperatures in the range 1.2 to 2.2 K and frequencies around 10^4 Hz.

(iv) Since the speeds of 1st and 2nd sound differ by approximately a factor of 10, there is increased dynamic range in any experiment simply by changing from a 1st sound measurement to that of 2nd sound.

(v) A very thin film ($\sim$ 100 Å) of superfluid is capable of supporting sound waves called 3rd sound. This mode has great potential for probing surface transport phenomena in porous media (see below).

The attenuation and the dispersion of the two acoustic modes of He II in a fully saturated porous medium can be calculated in terms of the dynamic permeability $\tilde{k}(\omega)$ assuming that the viscous drag on the normal fluid is the dominant effect [3]. In figure 3 we see the results of a comparison of the calculated dispersion and attenuation of 1st and of 2nd sound against the measured values [1]. The only input parameters

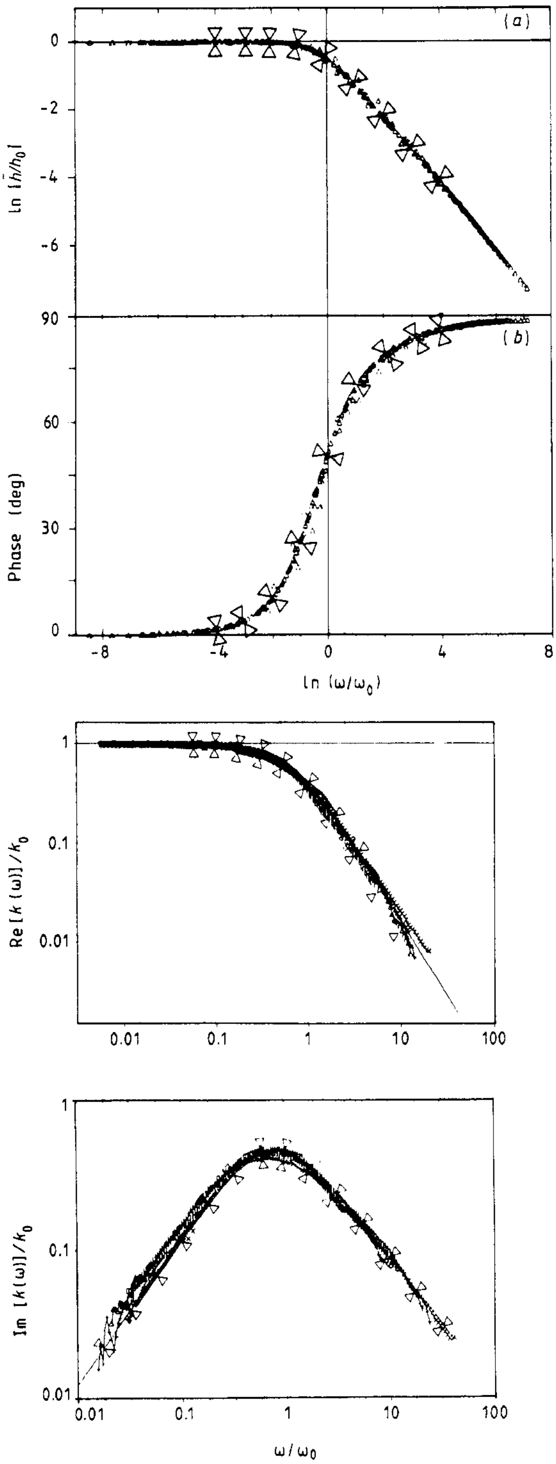

Figure 2. A comparison of the dynamical permeability predicted by equation (4) (the 'bow-ties') against (*a*) numerical calculations and (*b*) experimental measurements in a variety of geometries. After [6].

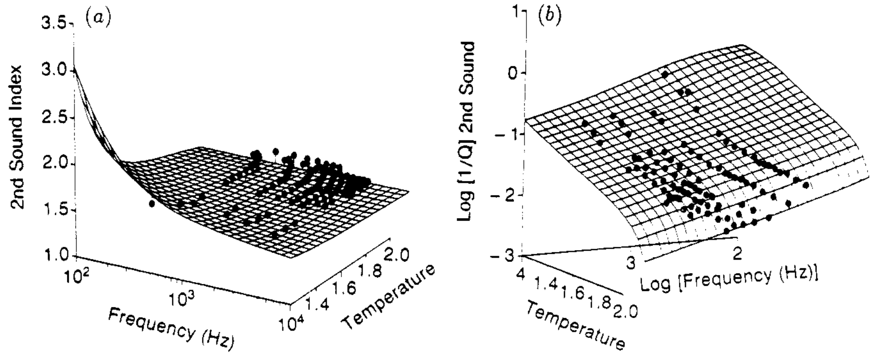

Figure 3. (a) index of refraction and (b) specific attenuation of the 2nd sound mode in a porous medium, QF-20 (registered trademark), manufactured by the Ferro Corporation. The surface is the theoretical prediction based on the model dynamic permeability, equation (4) using the following values of the four parameters: ϕ = 40.2%, k = 16.8 darcys, α = 1.89, Λ = 19.0 μm; all but Λ were determined by independent measurements. There is a vertical line drawn from each datum to the surface. After [1].

(aside from the He II properties) are those listed in the figure. Without going into detail here, we note that in the high-frequency limit of the theory the speeds of the two modes in the porous medium, $[V_i : i = 1, 2]$ are related to the bulk speeds of 1st and 2nd sound, C_1 and C_2, by $V_i = C_i/\sqrt{\alpha_3}$; the specific attenuations of these modes are directly related to Λ, viz: $1/Q_i \propto \delta/\Lambda$. Thus we have a sensitive acoustic means of determining the two electrical parameters, α_3 and Λ.

4. Biot theory of acoustics in fluid-saturated porous media

Suppose the porous medium is now saturated with an ordinary viscous fluid (e.g. water) whose compressibility is comparable to that of the solid. The Biot theory of acoustics was developed to handle this situation [7]; since both solid and fluid phases can move independently of each other, though their motions are coupled, the Biot theory predicts two longitudinal modes of propagation as well as the usual transverse (shear) mode. The theory has tremendous predictive power in that all of the input parameters can be measured independently [7]. One such input is the dynamic permeability, $\tilde{k}(\omega)$, which, as we have seen, can be deduced from superfluid acoustics measurements. The results of such a comparison, theory versus experiment, are presented [1] in figure 4. There are no adjustable parameters in the theory, all of them having been measured directly.

5. 4th sound and healing length effects

If the pores are sufficiently small that the normal fluid cannot move (or if the frequency is sufficiently low that the viscous skin depth is greater than Λ), one has 4th sound, a mode whose temperature dependence is well understood both theoretically and experimentally. If the pores are small enough there comes a point where the walls of

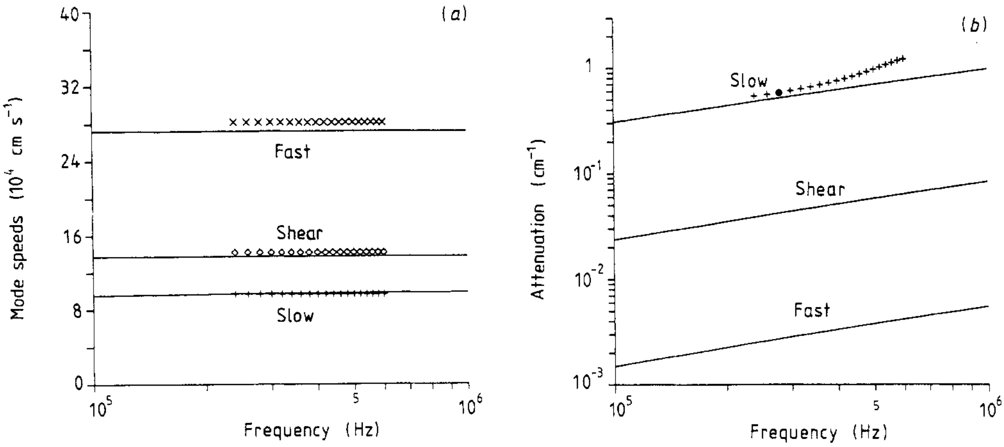

Figure 4. (*a*) Speeds and (*b*) attenuation of the fast, slow, and shear acoustic modes in a water-saturated sample of fused glass beads. The full curves are calculated from the Biot theory of acoustics in porous media wherein all the input parameters have been measured independently. The symbols are the measured speeds. After [1].

the porous medium act to reduce the superfluidity over a distance away from the wall called the healing length, $\xi_{\rm H}(T)$. The variation of healing length with temperature has been deduced, approximately, from 3rd sound measurements; it increases from 3.6 Å to $\sim$ 10 Å as the temperature is increased from 1.2 K although it presumably diverges at T_λ. Within the context of this treatment of healing length effects, the speed of 4th sound is [2]

$$V_4^2 = \frac{C_4^2}{\alpha_3}\left(1 - \frac{2\xi_{\rm H}}{\Lambda}\right). \tag{5}$$

where $C_4^2 \equiv (\rho_{\rm s}/\rho)C_1^2$ is the 'bare' speed of 4th sound and α_3 and Λ have their usual meanings. Thus, the temperature dependence of the speed of 4th sound in these small-pore media may prove to be a useful way to probe them; $\xi_{\rm H}(T)$ is a tunable yardstick.

6. 3rd sound

3rd sound is a mode which propagates in a thin film of superfluid $\sim$ (50–150) Å with a speed, C_3, which is dependent on $\rho_{\rm s}/\rho$ and on film thickness. If such a film coats the internal walls of a porous medium, the speed is renormalized to [1]

$$V_3 = C_3/\sqrt{\alpha_2}\,. \tag{6}$$

A value of α_2 in a porous medium consisting of a packed powder has been successfully deduced in this way (see [1]).

7. Shaly sands

We now return to a practical application of the electrical problem upon which we have based the foregoing discussion: electrical conduction in brine-saturated porous media in which there is appreciable conduction through the electrolytic double layer which coats the internal walls of the pore space. Typically this occurs in sedimentary rocks containing clay minerals. This is a complicated problem involving cation and anion motion, each of which moves through diffusion as well as conduction. Here, we point out that in these systems it is not known how the surface conductance, Σ_s, depends on brine salinity. By measuring the relevant geometrical parameters with the superfluid acoustic techniques sketched out above, it should become possible to deduce the value of Σ_s and its salinity dependence in real systems. A practical way of doing this involves the use of Padé approximant techniques [8].

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