

Some Macroscopic Considerations on Motions of Anisotropic Superfluids*

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We give a Herring-Kittel type of discussion of the nuclear-magnetic-resonance phenomena in the presumed anisotropic superfluid phase of liquid ^3He . The results agree with Leggett's derivation for high fields in an Anderson-Morel phase but differ fundamentally in other regimes. We observe that all observations so far published suggest that the "A" phase is of Anderson-Morel type (probably $L=1$), and the "B" phase of Balian-Werthamer type.

After the observation¹ of a shift of the nuclear-magnetic-resonance frequency in liquid ^3He in the "A" phase just below the presumed superfluid transition near 2.7 mK, Leggett² gave a sum-rule argument estimating the magnitude of the shift, and Anderson and Varma³ suggested a microscopic mechanism which gave the right magnitude (but turned out to be incorrect in most cases). Below the lower, "B" transition, the shift vanishes, although Leggett's sum-rule estimate does not vanish in any reasonable case.

We give here a more direct, macroscopic treatment on a semiphenomenological level, which (a) suggests that Leggett's conclusions are not quite correct; (b) agrees with the observations if the sequence of phases is normal \rightarrow Anderson-Morel⁴ superfluid (A) \rightarrow Balian-Werthamer⁵ superfluid (B) (as I suspect to be the case on other grounds as well); (c) predicts a number of other phenomena, including a very low-frequency resonance on the order of 1 Hz in the A phase, which may be responsible for the observed anomalous fluctuations; (d) is susceptible of modification to estimate dispersion of collective excitations.

The basis of our argument is the observation that there are at least three distinct directions (or systems thereof) which characterize the states of the anisotropic superfluid: the nuclear magnetization \vec{M} , the reference system for the spins of the condensed pairs (which we will schematize as a single vector \hat{k}_χ), and the reference system for the orbital motion of the pairs, which again we will schematize by a single vector \vec{L} .

These three systems are connected by a set of interaction free energies which are very different between the two cases of Anderson-Morel and Balian-Werthamer states (AM versus BW). In AM states the spins are paired with $M_s = \pm 1$ along \hat{k}_χ which is in fact a unique vector; while there is a total orbital angular momentum⁴ of order

$L \sim \hbar\Delta/E_F$ per atom pair, where Δ is the gap and E_F the Fermi energy (actually independently for each spin). Aside from weak and unknown boundary forces, \vec{L} is coupled to \hat{k}_χ by dipolar interaction energies of order

$$V_D \simeq (\Delta^2/E_F^2)n\mu^2 \sim 10^{-28} \text{ erg/atom.} \quad (1)$$

Since the anisotropic density correlation reproduces the angular shape of Δ , in the AM state for $\vec{L}=1$ the preferred orientation will be $\vec{L} \perp \hat{k}_\chi$, this providing the lowest dipolar energy. Thus

$$E(\hat{k}_\chi, \vec{L}) \simeq V_D \sin^2[\cos^{-1}(\hat{k}_\chi \cdot \vec{L}/k_\chi L)]. \quad (2)$$

The magnetization is coupled to \vec{L} only by *very* weak dipolar forces; but it is strongly coupled to \hat{k}_χ by the rather large anisotropy of the susceptibility,

$$E(M) = \frac{1}{2}\vec{M} \cdot (\chi)^{-1} \cdot \vec{M}, \quad (3)$$

where

$$\begin{aligned} \chi^{-1}(\parallel \hat{k}_\chi) &= \chi_N^{-1}, \\ \chi^{-1}(\perp \hat{k}_\chi) &= \chi_\perp^{-1} > \chi_N^{-1} \end{aligned} \quad (4)$$

(probably by a large factor because of susceptibility enhancement). Thus there is an energy

$$E(\vec{M}, \hat{k}_\chi) = -\frac{1}{2}\Delta(\chi^{-1})(\vec{M} \cdot \hat{k}_\chi)^2. \quad (5)$$

In the BW case, χ is isotropic so that no energy couples M and the other vectors, while the \vec{L} system has no net angular momentum. There is still a coupling of the same order as (1) between the \vec{L} and \hat{k}_χ systems, but of a more complicated form.

The second point of our treatment is to set

$$d\vec{J}/dt = \text{torque} = \partial E/\partial \theta, \quad (6)$$

where \vec{J} is any angular momentum.⁶ In doing this we neglect fluctuating relaxation torques, assuming all relaxations either too fast or too slow

to be relevant.

In the AM case the torques acting on \vec{L} are very weak and we may, at NMR frequencies, assume \vec{L} fixed. \hat{k}_χ , however, is coupled both to \vec{L} and to \vec{M} , and its direction will be determined by the stronger coupling constant. Assuming, as will be the case for resonance experiments, that \vec{L} , \hat{k}_χ , and \vec{M} are all nearly in their optimum directions, we can eliminate \hat{k}_χ and replace it by an effective \vec{M} - \vec{L} coupling energy

$$\frac{1}{2}(V_A)_{\text{eff}}(\Delta\theta_{ML})^2, \quad (7)$$

where

$$(V_A)_{\text{eff}} = \frac{\frac{1}{2}V_D M^2 \Delta(\chi^{-1})}{V_D + \frac{1}{2}M^2 \Delta(\chi^{-1})}, \quad (8)$$

and $\Delta\theta_{ML}$ is the angle of misorientation of \vec{L} and \vec{M} from their optimum relative orientation which is probably 90° , as we remarked.

The field at which

$$V_D = \frac{1}{2}M^2 \Delta(\chi^{-1}),$$

i.e.,

$$H_C = V_D / \chi^2 \Delta(\chi^{-1}), \quad (9)$$

is a transition field between the regimes

$$(V_A)_{\text{eff}} = V_D \quad (\text{high } H), \quad (10a)$$

and

$$(V_A)_{\text{eff}} = \frac{1}{2}M^2 \Delta(\chi^{-1}) \quad (\text{low } H), \quad (10b)$$

and is roughly T independent, since both the anisotropy of χ and V_D are $\propto \Delta^2$. This is presumably the field below which the resonance shift can no longer be observed,⁷ and represents a maximum possible shift on the field axis.

Above this field, it is easy to calculate the NMR shift by using $dM/dt = \text{torque}$, and we get

$$\Delta\omega/H = V_D / \chi H^2, \quad (11)$$

which is the same expression given by Leggett; but the shift is much smaller for $H < H_C$ and does not agree with Leggett's. We have assumed that because no angular momentum is associated with \hat{k}_χ it can follow torques infinitely rapidly (or at least at approximately the gap frequency). Only the last conclusion depends on this remark, which mainly conceals my ignorance of the dynamics of \hat{k}_χ . If we think of the spin system as two strongly coupled, macroscopic, opposite \vec{S} vectors, as in an antiferromagnet, this remark is essentially true.

In the BW state, the \hat{k}_χ system is fixed to the

\vec{L} system by dipolar interactions, but neither exerts any appreciable torque on the \vec{M} system, since χ is not anisotropic relative to \hat{k}_χ or, *a fortiori*, \vec{L} . Thus we predict *no* appreciable resonance shift in this case because there is no extra torque on \vec{M} , again in direct contradiction to Leggett. We do not predict any resonant absorption for $\Delta H \parallel H$, merely ordinary relaxation plus high-frequency absorption, because no torques are exerted on any internal systems. Again we disagree fundamentally with Leggett. It is well known that sum-rule arguments, as Leggett remarks, do not necessarily locate resonances exactly (one is always at the mercy of nonresonant contributions to χ'' .) Thus we do not consider these disagreements serious. The absence of shift for Balian-Werthamer states affords a reasonable explanation for the B transition and a strong clue to its nature.

Finally, we note that at a sufficiently low frequency \vec{L} can precess relative to the coupled \vec{M} - \hat{k}_χ systems. The \vec{L} - \vec{M} coupling via \hat{k}_χ and V_D corresponds to a precession frequency

$$V_D/L \sim \Delta n \mu^2 / E_F \hbar \sim 10 \text{ Hz}, \quad (12)$$

but L is confined only in a plane perpendicular to \vec{M} and its precession will be elliptical at some lower frequency. If L is pinned at the boundaries, the low modes of motion will correspond to orbital spin waves of wavelength of the order of the sample size. "Orbit waves," again, can be estimated using the torque equation and the reasonable estimate of the spatial coupling energy

$$E_{sp} = (\Delta^2 n / E_F) (\xi \nabla L / L)^2 \quad (13)$$

which, in the absence of dipolar interaction, would give a spin-wave frequency of about 10^{-2} for 1 cm wavelength. "Orbit waves" of high frequency will have frequencies

$$\omega \simeq (\hbar k^2 / m) E_F / \Delta \simeq 10^{-2} k^2 \text{ Hz}. \quad (14)$$

At low frequencies, for the elliptically precessing modes we must take the square root of the product of (12) and (14),

$$\omega \simeq 0.3k, \quad (15)$$

which for centimeter wavelengths gives frequencies of the order of the anomalous fluctuations observed by Osheroff.⁶

The above picture is very qualitative but I believe contains many of the relevant physical phenomena. We see that these phenomena are extremely complicated because of the internal com-

plexity of the anisotropic superfluid state, and it may be unlikely that all aspects have been correctly treated here.

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⁵R. Balian and N. R. Werthamer, *Phys. Rev.* **131**, 1553 (1963).

⁶The treatment is in the spirit of the Herring-Kittel treatment of spin waves [C. Herring and C. Kittel, *Phys. Rev.* **81**, 869 (1951)].

⁷D. D. Osheroff, thesis, Cornell University, 1972 (unpublished).

Valence Band of Mg₂Sn Determined by Auger and Photoemission Spectroscopy

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It is shown that Auger spectroscopy can be used to determine the density of valence states in compound semiconductors. We determined the density of valence states by this method in Mg₂Sn and find it in basic agreement with those which we obtained by x-ray- and uv-induced photoelectron spectroscopy. A comparison is made with the calculated Mg₂Sn valence band. The Mg₂Sn valence band obtained by Auger spectroscopy (involving the *KL* transition in Mg) is contrasted with the analogous spectrum for Mg metal.

Measurements of valence-band densities of states have recently been carried out by a number of authors using electron-emission spectroscopy.¹⁻⁵ Depending on the way the emitted electrons were originally excited, either by x-rays or far-ultraviolet radiation, the experimental methods are referred to as x-ray-induced electron-emission spectroscopy (XPS) (also called ESCA) and uv-induced electron-emission spectroscopy (UPS). Measurements with both methods have resulted in a number of experimental valence-band densities of states for several metals and semiconductors.¹⁻⁶

Each of these methods has its advantages and drawbacks. These are caused mainly by differences of the depth of the surface layer which is sampled and by the effect of transition probabilities to the quasicontinuous final states. XPS measurements sample a deeper layer (~25 Å) than UPS (~5 Å) because of the higher velocity of the escaping electrons and are therefore more characteristic of the bulk properties: They should be preferred if simultaneously good resolution is obtained. On the other hand, with present experimental setups, the resolution is usually better

for UPS measurements for which the condition of the surface plays a more important role. The second effect manifests itself as a dependence of the "nominal" valence-band density of states on the energy of the exciting uv radiation. However, as long as valence-band spectra obtained with 21- and 41-eV photons are similar, it is reasonable to conclude that the effects of absorption and of escape probabilities on the "nominal" density of valence states are negligible.

In this Letter we compare the above methods with a third way to measure the valence-band density of states which, in conjunction with other high-energy spectroscopic measurements, offers wide possibilities for the analysis of the valence bands, especially in compound semiconductors. This method is based on the Auger effect,⁷ usually related to core states of atoms described by *WXY* (*W* denotes the core level where initially a hole is created by emission of an electron, *X* denotes the core level from which an electron is provided to fill the hole in the energetically deeper-lying *W* level, and *Y* the level from which another electron is expelled). If one of the two levels *X* or *Y* happens to be the valence band,