Observation of Magneto-Elastic Effects in Terbium Metal by Ferromagnetic Resonance at $\lambda = 2.5$–3 mm*

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Magneto-elastic effects recently included in theoretical studies of spin-wave spectra of heavy rare-earth metals have suggested that millimeter ferromagnetic-resonance (FMR) results in terbium metal should be reinvestigated. We present data here from a new, detailed study of FMR in single-crystal terbium metal. The data extend over a wider temperature range (13–240 K) than was previously reported, and at frequencies of 98 to 121 GHz. Angular studies were also made. The high-frequency FMR data are qualitatively the same as those reported earlier, and agree qualitatively with theoretical calculations from the model which assumes that elastic strains are "frozen" at the equilibrium (i.e., no spin-wave) configuration. Quantitative fitting of the theory to the data requires values of the anisotropy and magneto-elastic constants that are in reasonable agreement with the results of other studies. Our values are $P_2S = -18$ K/atom and $D\gamma = 1.7$ K/atom. The present paper provides a solution to the puzzling discrepancy which has existed for several years between our earlier high-frequency FMR results and Cooper's theoretical predictions.

I. INTRODUCTION

RECENTLY, magneto-elastic effects have been included in the Hamiltonian for the heavy rare-earth metals in a theoretical investigation by Cooper.[1] A discussion was given of the puzzling discrepancy among several experiments involving magnetic anisotropy in Tb metal. The dependence of resonance field on temperature predicted by Cooper did not agree qualitatively with earlier 3-mm ferromagnetic-resonance (FMR) studies on Tb.[2] This disagreement suggested that a new FMR study of Tb might help clarify the situation.

Here we present data from such a new, detailed study of FMR in single-crystal terbium metal, extending over a wider temperature range (13–240 K) than was previously reported. The present data also cover a wider range of frequencies, with resonances observed from 98 to 121 GHz. We also compare our data with Cooper's theory, which includes magneto-elastic effects.

II. THEORY

Cooper has thoroughly treated the temperature and field dependence of ferromagnetic resonance in the heavy rare earths.[1] We will here sketch only the main points leading to the results that we need to interpret our data.

First, the Hamiltonian for the spins in the absence of a magneto-elastic interaction and in the absence of an external field is

$$\mathcal{H} = -\sum_{i\neq j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j - \sum_i \{P_2 S_{i\zeta}^2 - \tfrac{1}{2}P_6^6 \times [(S_{i\xi}+iS_{i\eta})^6 + (S_{i\xi}-iS_{i\eta})^6]\}, \quad (1)$$

where J_{ij} is the exchange coupling constant, P_2 and P_6^6 are anisotropy constants, and S is the total (spin + orbital) angular momentum. The Holstein-Primakoff approximation gives a spin-wave dispersion

$$\hbar\omega(q) = S^{-1}\{[-2P_2S^2 + 2S^2J(0) - 2S^2J(q) - 6P_6^6S^6] \times [2S^2J(0) - 2S^2J(q) - 36P_6^6S^6]\}^{1/2}, \quad (2)$$

where the $J(q)$ are the Fourier coefficients of J_{ij}.

The microwave fields couple with spin waves for $q \sim 1/\delta$, where δ is the penetration depth of the microwave fields into the metal ($\delta \sim 10^{-6}$ m) and q at the edge of the Brillouin zone is $\sim 10^9$ m^{-1}. We are thus measuring $\omega(0)$ to a good approximation.

The temperature dependence of the anisotropy constants is given by the theory of Callen and Callen[3] as $\hat{I}_{l+1/2}[\mathcal{L}^{-1}(\sigma(T))]$, where l is the order of the anisotropy, $\hat{I}_{l+1/2}$ is the hyperbolic Bessel function of the order $l+\frac{1}{2}$ divided by that of order $\frac{1}{2}$, $\mathcal{L}^{-1}$ is the inverse Langevin function, and $\sigma(T)$ is the ratio of the magnetization at temperature T to that at $T=0$. We found that using the low-temperature approximation for $\hat{I}_{5/2}$ gave considerably different values for the resonance field than did the true values of $\hat{I}_{5/2}$, so the latter were used exclusively in the computations.

Thus we arrive at

$$\hbar\omega(0) = [(-2P_2S\hat{I}_{5/2}/\sigma - 6P_6^6S^5\hat{I}_{13/2}/\sigma) \times (-36P_6^6S^5\hat{I}_{13/2}/\sigma)]^{1/2}, \quad (3)$$

where it is understood that $\omega(0)$ stands for $\omega(q=0)$ at temperature T and, where appropriate, at applied field $\mathbf{H}$. In the presence of a magnetic field applied along a hard axis this becomes

$$\hbar\omega(0) = [(-2P_2S\hat{I}_{5/2}/\sigma + 6P_6^6S^5\hat{I}_{13/2}/\sigma + g\mu_BH) \times (36P_6^6S^5\hat{I}_{13/2}/\sigma + g\mu_BH)]^{1/2}, \quad (4)$$

where g is the Landé factor and μ_B is the Bohr magneton. This equation is valid if the field is sufficient to align the magnetization along the hard axis, i.e., since $P_6^6 < 0$, we require that the second factor under the radical be

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[1] B. R. Cooper, Phys. Rev. **169**, 281 (1968); in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1968), Vol. 21.

[2] J. L. Stanford and R. C. Young, Phys. Rev. **157**, 245 (1967).

[3] E. Callen and H. B. Callen, Phys. Rev. **139**, A455 (1965).

positive. Notice at this point that by applying a field in the hard direction we can lower the spin-wave gap from its value in zero field. In fact, we can reduce it to zero for $H=-36P_6{}^6S^5\hat{I}_{13/2}/\sigma\mu_B$. Resonance occurs when the experimental frequency $\omega_{rf}=\omega(0)$.

Now, when magneto-elastic coupling between spin and lattice is taken into account, two more terms are added to the spin Hamiltonian: $\mathcal{H}_E$ and $\mathcal{H}_M$, where $\mathcal{H}_E$ is the elastic energy associated with the strains and $\mathcal{H}_M$ is the interaction between spins and strains. The equilibrium configuration of the strains for a given magnetization is found by minimizing the free energy with respect to the strains.

The question that arises at this point is: What happens to the strains in the presence of a spin wave? Do they follow the instantaneous direction of magnetization (thus keeping the magneto-elastic energy at a minimum), or do the strains remain frozen at their equilibrium value while the magnetization changes? The latter is the "frozen lattice" model proposed by Turov and Shavrov.[4] Both cases are treated quantitatively by Cooper.

In the first case, since the lowest-order magneto-elastic energy has cylindrical symmetry, there is no first-order contribution to the spin-wave gap. Treating the magneto-elastic energy to second order gives a term having hexagonal symmetry, but with a temperature dependence different from that of the crystal field anisotropy term $P_6{}^6$. The resulting expression for the spin-wave energy is

$$\hbar\omega(0)=\{[-2P_2S\hat{I}_{5/2}/\sigma+6(P_6{}^6S^5\hat{I}_{13/2}/\sigma+C\hat{I}_{5/2}\hat{I}_{9/2}/\sigma)+g\mu_BH][36(P_6{}^6S^5\hat{I}_{13/2}/\sigma+C\hat{I}_{5/2}\hat{I}_{9/2}/\sigma)+g\mu_BH]\}^{1/2}, \quad (5)$$

where C is a magneto-elastic constant evaluated by Cooper[1] as approximately -0.16 K/atom, which is comparable to $P_6{}^6$. Since the magnetostriction measurements include the effects of both $P_6{}^6$ and C, Cooper has calculated the spin-wave spectrum for two extreme cases: all hexagonal anisotropy due to $P_6{}^6$, $C=0$ ("unstrained hexagonal anisotropy"), and all hexagonal anisotropy due to C, $P_6{}^6=0$ ("hexagonal anisotropy due to strain"). The temperature dependence of these two cases is not very different, and both show a rise in resonance field at low temperature.

We now consider the case of the elastic strains frozen at the equilibrium position. Here the lowest-order magneto-elastic energy does not vanish, and its effect on the spin-wave energy as calculated by Cooper[1] is

$$\hbar\omega(0)=\{[-2P_2S\hat{I}_{5/2}/\sigma+6P_6{}^6S^5\hat{I}_{13/2}/\sigma+g\mu_BH+D^\gamma(\hat{I}_{5/2})^2/\sigma][36P_6{}^6S^5\hat{I}_{13/2}/\sigma+g\mu_BH+2D^\gamma(\hat{I}_{5/2})^2/\sigma]\}^{1/2}, \quad (6)$$

where $2D^\gamma=4.35$ K/atom (Cooper's evaluation from other data). What is significant here is the positive sign of D^γ. If $|36P_6{}^6S^5|\leq 2D^\gamma$, then the spin-wave energy cannot be reduced by an external field along a hard direction, as was the case when $D^\gamma=0$. The resonance field as a function of temperature will show a monotonic decrease with decreasing temperature until it is impossible to satisfy the resonance condition. Experimentally, this is what we observe in Tb at high microwave frequencies, except that below about 4 kOe the field is insufficient to align the domains, and no resonance is seen. (We do not know how to calculate the effect on microwave absorption of a multidomain specimen.)

III. EXPERIMENTAL PROCEDURES

The experimental configuration was the same as that described in Ref. 2: disk-shaped sample with [0001] normal. The applied magnetic field H lay in the plane of the disk and, except for the angular and polarization studies, was along a hard direction of magnetization (a axis) and perpendicular to the microwave magnetic field. The high-frequency experimental apparatus is essentially the same as that reported earlier. One exception is that much of the present data were obtained with a "magic T" instead of a directional coupler used in the microwave spectrometer. This resulted in enhanced signal-to-noise quality of the data. At the higher frequencies, 108–121 GHz, a Phillips klystron was used. Typical power outputs were a few milliwatts. Mounting the temperature-measuring thermocouple closer to the sample cavity resulted in more accurate determination of sample temperatures. One fixed temperature, that of a dry-ice–acetone mixture, was employed to provide a fixed point in the region of large temperature dependence of resonance field. In this instance we could be sure that the sample and sample-holder were at a uniform temperature, and the good agreement of the data taken at this temperature and in this manner with those taken the usual way (small heat leak to liquid nitrogen or helium bath) assured us that there was no large unknown thermal gradient between sample and temperature-monitoring device.

One of the samples used is the same as reported in Ref. 2. It was reelectropolished several times during the course of the experiment. No change in resonance behavior was noted. Another crystal, having a residual resistivity ratio of approximately 50, twice that of the original sample, was prepared. It had a diameter of 5.9 mm and thickness of 0.8 mm, compared with 9.5 and 2.3 mm, respectively, for the first sample. After studying the temperature dependence of the resonance at 98.2 GHz in this specimen, its thickness was further reduced by abrasion and electropolishing to 0.4 mm, and the microwave resonance studied again. Within the scatter of the data, the resonance field at a given temperature was found to be independent of sample thickness.

[4] E. A. Turov and V. G. Shavrov, Fiz. Tverd. Tela **7**, 217 (1965) [English transl.: Soviet Phys.—Solid State **7**, 166 (1965)].

In the earlier experiment, the sample was glued from the back to the cavity tuning mechanism (see Fig. 2 of Ref. 2). For most of the present high-frequency investigation, the front of the sample was held against a concentric ring which was fastened to the tuning mechanism. This change in mounting allowed the sample to constrict (due to magnetostriction) without appreciably changing the resonant frequency of the cavity. This was less important with the thinner samples, and they were mounted in the manner described in Ref. 2.

The polarization study was carried out by rotating the dc magnetic field in the plane of the specimen, while keeping the microwave polarization constant. At temperatures above 170 K no shift in peak position was observed, indicating the validity of this procedure.

IV. RESULTS AND DISCUSSION

Figure 1 shows tracings of representative experimental curves in the 2.5–3-mm region. The temperature dependence of the fields at which absorption maxima occur is given in Fig. 2. Cooper has predicted the dependence of resonance field on temperature for Tb metal at 100 GHz for the three cases previously discussed, namely, unstrained hexagonal anisotropy, hexagonal anisotropy due to strain, and the frozen-lattice model. These are shown as curves A–C, respectively. The solid line is our calculation from Cooper's expression for the frozen-lattice case, but with parameters fitted at two points on the experimental curve. These fitted values were (in Cooper's notation) $P_2S=-18$ K/atom and $D^\gamma=1.7$ K/atom,

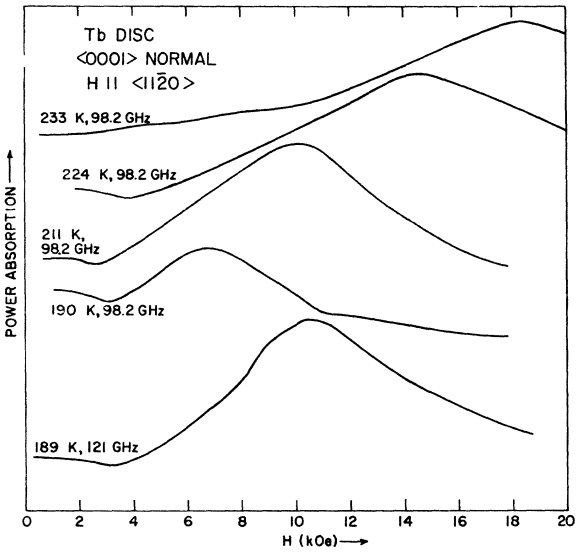

FIG. 1. Tracings of experimental curves of microwave power absorption versus applied magnetic field. H_{rf} is perpendicular to the applied field H, both lying in the plane of the sample. The scales for the various curves are not all the same. The absolute magnitude of the absorption is a few percent, after enhancement by the cavity Q of 1000–2000.

or, using Rhyne and Clark's notation, $K_2=3.1\times10^8$ erg/cc and $D^\gamma=7.3\times10^6$ erg/cc.[5]

Marsh and Sievers[6] have measured the spin-wave energy gap in polycrystalline Tb as a function of temperature by means of far-infrared absorption. Their data are also consistent with the frozen-lattice model. They assume a value for the axial anisotropy of

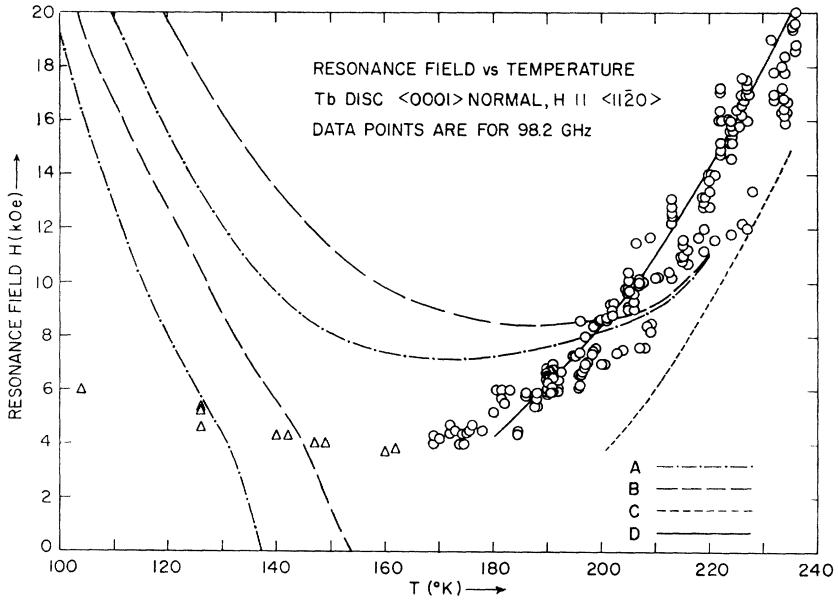

FIG. 2. Plot of the field values of absorption peaks versus sample temperature. Theoretical curves as follows: A, unstrained hexagonal anisotropy; B, hexagonal anisotropy due to strain; C, frozen-lattice model; D, frozen-lattice model with fitted parameters. Curves A and B calculated by Cooper for $f=100$ GHz; curves C and D calculated by present authors for $f=98.2$ GHz. The triangles represent absorption maxima believed to be associated with the onset of magnetostriction.

[5] J. J. Rhyne and A. E. Clark, J. Appl. Phys. **38**, 1379 (1967).
[6] H. S. Marsh and A. J. Sievers, J. Appl. Phys. **40**, 1563 (1969).

$K_2=4.5\times10^8$ erg/cc, taken from magnetization data of Rhyne *et al.*,[7] and determine $D^\gamma=8.3\times10^6$ erg/cc.

A word should be said about the method used to calculate the solid line in Fig. 2. Using expression (6), taken from Cooper's paper, the resonance field value was calculated[8] for various values of the magnetization. Then the magnetization data of Hegland *et al.*[9] for Tb were used to find the temperature at which this magnetization is achieved in an effective field equal to the applied field reduced by the demagnetizing field for the particular sample in question. A linear interpolation between adjacent temperatures in Hegland's data was used.

The triangles in Fig. 2 represent absorption maxima which we believe to be associated with the onset of magnetostriction in our crystal. The temperature dependence of the field values of this structure correlates well with magnetostriction data of Rhyne and Legvold[10] which were taken on one of the Tb disks used in our experiment. The resonance line was followed down in temperature until, between 160 and 170 K, it disappeared below the magnetostriction-onset structure at approximately 4 kOe. The resonance line was never observed to rise above this structure in the temperature range 13–160 K. The trailing (high-field) edge of the large linewidth was observed to decrease in field as the temperature was lowered in the range below 160 K, even though the absorption peak itself was not observed. (Evidently it would occur at a field value below that necessary for the onset of magnetostriction.) All our resonances were observed in fields higher than the magnetostriction-onset field. It is not clear to us how to apply the theory to a multidomain sample, as is the case below the onset of magnetostriction. For this reason, in plotting theoretical curves, we have not plotted any points below about 4 kOe, approximately the field at which domain alignment occurs.[11]

Angular studies near the lowest temperatures for which the absorption peak can be directly observed revealed no hexagonal anisotropy within the scatter of the experimental data; the resonance field was found to be independent of the field direction in the basal plane.

The "jump" in the resonance field at T_c reported previously[2] was not observed in the present investigation. The 19 kOe temperature-independent line also was not observed, confirming our earlier suspicion[2] that it was not related to a bulk phenomenon. Better positioning of the thermocouple which measures the sample temperature has led to more accurate H-versus-T data for temperatures above T_N than were previously reported. We believe, however, that some of the data scatter is still due to differences between sample temperature and the monitored temperature.

The linewidths are characteristically very broad (5–10 kOe), similar to our earlier results and to the large linewidths observed by Rossol and Jones[12] at 38 GHz in Dy.

Polarization studies showed that the amplitude of the absorption peak varies as the square of the sine of the angle between H_{rf} and H_{dc}, the expected dependence for spin-wave excitations.

For the samples of different thickness which were used in this experiment, the resonance field at a given temperature was independent of thickness, within the scatter of the data. The only change with thickness which would be predicted on the basis of the theory is due to a change in the demagnetizing fields inside the sample. The effect on the H-versus-T curve is small, and much less than the scatter of the data. The field at which magnetostriction onset occurred was somewhat lower for the thinner samples.

The resonance field at fixed temperature was found to increase with increasing frequency in the range studied, 98–121 GHz. Values of resonance field at 190 K were measured over the frequency range. Scatter in the data was too great to determine a power-law dependence, but it was consistent with $H\propto\omega^2$.

V. SUMMARY AND CONCLUSIONS

The results of a careful study of the microwave power absorption at $\lambda=2.5$–3 mm in single crystals of high-purity Tb metal have been presented. The investigation included a wider temperature range than previously reported, a study of the effect of different sample thicknesses, and angular studies where the applied field was rotated in the basal plane of the sample. Polarization studies confirmed that the amplitude of the absorption peaks obeyed the dependence expected for spin-wave excitation.

The temperature dependence of the resonance field at 98.2 GHz was compared with the theory of Cooper for hexagonal anisotropy due to strain and for the case of unstrained hexagonal anisotropy. The experimental data fail to exhibit the rise of resonance field at low temperature required by either theory.[13] The calcu-

[7] J. J. Rhyne, S. Foner, E. J. McNiff, Jr., and R. Doclo, J. Appl. Phys. **39**, 892 (1968).

[8] The first factor under the radical in Eq. (6) should read $H+4\pi M$ instead of H, because of high-frequency demagnetizing effects. There should also be a term, omitted from our calculations, to account for static demagnetizing effects. For our samples this term introduces a correction small compared with the scatter in the data.

[9] D. E. Hegland, S. Legvold, and F. H. Spedding, Phys. Rev. **131**, 158 (1963).

[10] J. J. Rhyne and S. Legvold, Phys. Rev. **138**, A507 (1965).

[11] As pointed out above, for a single-domain sample the frozen-lattice theory predicts the existence of a temperature below which the resonance condition can not be met at any value of field.

[12] F. C. Rossol and R. V. Jones, J. Appl. Phys. **37**, 1227 (1966).

[13] We should stress that in order to match the value of $\hbar\omega(0)$ found at 90 K in Tb by neutron scattering, for the calculations omitting frozen-lattice effects, Cooper had to adopt a value $36P_6{}^6S^5=-12.55$ K, i.e., about 3.7 times the experimental static value. Curves A and B of our Fig. 2 are then Cooper's calculated curves on this basis. On the other hand, when frozen-lattice effects were included, Cooper could closely fit the neutron scattering value of $\hbar\omega(0)$ using the various independently measured experimental anisotropy and elastic constants.

lations were then performed using Cooper's theory for the frozen-lattice model. Good qualitative agreement between theory and experiment resulted. Quantitative agreement is obtained if the values $P_2S=-18$ K/atom and $D^\gamma=1.7$ K/atom are chosen for the twofold anisotropy[14] and magneto-elastic constants.

[14] This value appears to be in satisfactory agreement with the neutron diffraction results of H. Bjerrum Møller, J. C. Gylden Houmann, and A. R. Mackintosh [Phys. Rev. Letters **19**, 312 (1967)], for P_2S.

In conclusion, we believe that this paper reconciles the earlier discrepancy between experiment and theory for ferromagnetic resonance in Tb at high microwave frequencies.

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Rigorous Boson Formulation for Calculating Time-Dependent Thermal Properties of Localized Spin Systems*

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We outline a method for rigorously calculating the time-dependent as well as the static thermal properties of localized spin systems from those of an ordinary many-boson system. Our method retains the advantages of both the Holstein–Primakoff and the Dyson–Maleev transformations without having their main disadvantages. The spin operators and the boson Hamiltonian are all finite series in the boson creation and annihilation operators. Our boson Hamiltonian is Hermitian. This method establishes a rigorous correspondence between the thermal properties of the spin-$\frac{1}{2}$ isotropic Heisenberg model and those of a hard-core boson system with only two-body interactions.

1. INTRODUCTION

THE methods available for the calculation of the thermodynamic properties of spin systems do not, as yet, seem to be as advanced as the methods[1,2] already developed for treating the thermodynamic properties of many-body systems of bosons or fermions. The reason for this is that the spin-operator commutation rules are more complicated than the boson or fermion commutation rules. In two famous papers in 1956,[3] Dyson used the idea of establishing a correspondence between a given spin system and a boson system, which then could be used to calculate thermodynamic properties of spin systems if the corresponding properties of the boson system could be calculated. He then proceeded to use this correspondence to calculate some of the static (nontime-dependent) properties of the Heisenberg ferromagnet.

Dyson's work was later extended in an effort to calculate the spin Green's functions which would, of course, give one a means of calculating the time-dependent as well as the static thermodynamic properties of spin systems.[4,5]

This approach, however, is only one of several that have been attempted. The usual starting point with these theories is to establish a transformation from the spin operators to a set of boson operators, the two most often used being the Dyson–Maleev[6] and the Holstein–Primakoff[7] transformations. The Dyson–Maleev transformation leads to a "boson system" described by a non-Hermitian "Hamiltonian." The Holstein–Primakoff transformation leads to a boson system with a Hermitian Hamiltonian which is an infinite series in the boson operators. The objections to using these transformations are obvious.

The object of this paper is to present a theory whereby the time-dependent as well as the static properties of a spin system could, in principle, be calculated by means of the application of ordinary many-body boson theory to a system of bosons described by a finite-series Hermitian Hamiltonian. The procedure we have in mind is the following: In order to calculate a given thermodynamic property of the spin system one must, in general, calculate some kind of time-dependent correla-

* Research sponsored by the U. S. Atomic Energy Commission under contract with Union Carbide Corporation.
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[2] L. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (W. A. Benjamin Inc., New York, 1962).
[3] F. J. Dyson, Phys. Rev. **102**, 1217 (1956); **102**, 1230 (1956).
[4] R. A. Tahir-Kheli and D. ter Haar, Phys. Rev. **127**, 95 (1962).
[5] R. Silberglitt and A. B. Harris, Phys. Rev. **174**, 640 (1968).
[6] S. V. Maleev, Zh. Eksperim. i Teor. Fiz. **33**, 1010 (1957) [English transl.: Soviet Phys.—JETP **6**, 776 (1958)].
[7] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1941).