neglected here. ²⁴ For $\eta \sim 10^4$ or 10^5 eV, the result is independent of η . One sees that the deviations from Bloch's formula range from about 3% for iron up to 20% for Z = 114. The deivations for negative Z are much less, probably because when the cross section is expanded as a series in Z, it becomes an alternating series over most of the energy range for negative Z. Figure 9 shows $(1/Z^2)(dE/dx)$ when the Bloch correction [terms containing Ψ in Eq. (3)] is omitted. The energy loss of positively charged nuclei exceeds that of negatively charged nuclei by up to 25% for $Z = \pm 114$.

In Fig. 10 we have plotted $[(dE/dx) - (dE/dx)_B]/(dE/dx)_B = D$ as a function of *I* for various *Z*'s and β 's where $(dE/dx)_B$ is calculated using $(d\sigma/dT)_{FB}$ in Eq. (2). The deviation *D* from the Bloch for-

¹N. Bohr, Phil. Mag. <u>25</u>, 10 (1913).

²H. A. Bethe, Ann. Physik 5, 325 (1930).

³E. J. Williams, Proc. Roy. Soc. (London) <u>139</u>, 163 (1933).

⁴F. Bloch, Ann. Physik <u>16</u>, 285 (1933).

[°]N. F. Mott, Proc. Cambridge Phil. Soc. <u>27</u>, 553 (1931). ⁶H. A. Bethe and J. Ashkin, *Experimental Nuclear Physics*, edited by E. Segre (Wiley, New York, 1953), Vol. 1,

Chap. 2; and U. Fano, Ann. Rev. Nucl. Sci. <u>13</u>, 1 (1963). 7 Reference 4, Eqs. (51) and (71).

⁸E. J. Williams, Rev. Mod. Phys. <u>17</u>, 217 (1945).

⁹E. A. Uehling, Ann. Rev. Nucl. Sci. <u>4</u>, 315 (1954).

¹⁰The idea of applying the Mott cross section to close collisions was suggested by Fowler *et al.* [Proc. Roy. Soc. (London) <u>A318</u>, 1 (1970)] in connection with charge identification of heavy nuclei from emulsion tracks.

¹¹N. Sherman, Phys. Rev. <u>103</u>, 1601 (1956).

 12 J. A. Doggett and L. V. Spencer, Phys. Rev. <u>103</u>, 1597 (1956).

 13 A similar but not identical integral involving the Mott cross section has been calculated by G. S. Khandelwal and E. Merzbacher, Phys. Rev. <u>130</u>, 1822 (1963), in a different context.

¹⁴J. W. Motz, Haakon Olsen, and H. W. Koch, Rev.

mula is a slowly increasing function of I and thus absorber atomic number z. This figure only applies when the last two terms in Eq. (3) can be neglected. The density effect would in general decrease the contribution of Eq. (3) to the totalenergy loss and thus increase the deviation D. We do not intend to give a comprehensive tabulation of results for various media but only to show that, in general, for relativistic particles with $Z \gtrsim 20$ the deviations from the Born approximation are significant and should be included in energy-loss calculations.

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Mod. Phys. <u>36</u>, 881 (1964).

¹⁵V. Z. Jankus, Phys. Rev. 90, 4 (1953).

¹⁶Bloch has apparently not included the correct relativistic correction in Eq. (65) of Ref. 4. This explains the discrepancy between Bloch's final result [Eq. (71), Ref. 4] and that quoted by L. C. Northcliffe, Ann. Rev. Nucl. Sci. 13, 67 (1963), Eqs. (4) and (6).

¹⁷G. S. Khandelwal, Nucl. Phys. <u>A116</u>, 97 (1968).

¹⁸R. M. Sternheimer, Phys. Rev. <u>164</u>, 349 (1967). ¹⁹See, S. K. Allison and S. D. Warshaw, Rev. Mod.

Phys. 25, 799 (1953); U. Fano, Ref. 6.

²⁰J. C. Ashley, W. Brandt, and R. H. Ritchie, Bull.
Am. Phys. Soc. <u>15</u>, 1338 (1970). See also G. Basbas *et al.*, Phys. Letters <u>34A</u>, 277 (1971).
²¹V. B. Semikoz, Zh. Eksperim. i Teor. Fiz. Pis'ma

²¹V. B. Semikoz, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiya <u>9</u>, 534 (1969) [Sov. Phys. JETP Letters <u>9</u>, 324 (1969)].

 22 J. E. Turner, NAS-NRC Publication No. 1133, Report 6 (unpublished). This value for *I* is given by Eq. (5) of Ref. 18.

²³J. W. Epstein *et al.*, Nucl. Instr. Methods <u>95</u>, 77 (1971).

²⁴R. M. Sternheimer, Phys. Rev. <u>88</u>, 851 (1952); Ref. 22.

PHYSICAL REVIEW A

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NMR in Nondilute Solid ³He-⁴He Mixtures

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An explanation of the anomalous NMR specific heat first seen by Garwin and Reich in nondilute solid ${}^{4}\text{He}{}^{-3}\text{He}$ mixture is given in terms of the energy of interaction of ${}^{4}\text{He}{}^{-4}\text{He}{}^{-4}$.

The purpose of this note is to put forth a tentative suggestion for the explanation of some of the unusual NMR data on ${}^{3}\text{He}{}^{-4}\text{He}$ mixtures. The data to which the explanation is addressed is that of Garwin and Reich, 1 Bernier and Landesman, 2 and Reich and Yu³ which suggests the existence of an anomalous specific heat due to the presence of ⁴He impurities at concentrations up to 1% in ³He. There are two things in the data that must be accounted for: (i) the basic interaction among the excitations in the system that leads to the observed T_1 and (ii) the number, magnitude, and kind of energy reservoirs that take the rf energy from the Zeeman system and which feed it through the basic interaction to the phonons. Guyer and Zane⁴ have proposed that the mass fluctuation wave-phonon interaction is observed in the T_1 experiments on dilute mixtures. The careful T_1 experiments of Giffard and Hatton⁵ and Giffard⁶ (the Oxford experiments) have provided reasonable confirmation of the essential features of their proposal. The experiments of Garwin and Reich, Bernier and Landesman, and Reich and Yu are in the same temperature range as the Oxford experiments, but they go to higher concentrations of ⁴He impurities. These experiments see a T_1 with many features similar to those studied in the Oxford experiments (principally a T^{7-8} temperature dependence) and in addition they see a specific-heat anomaly. In the case of Bernier and Landesman the anomaly is observed for 60 ppm $\leq x_4 \leq 2800$ ppm at v = 20.0 cm³/mole; about a factor of 2 too much specific heat is seen at 1000 ppm. No excess specific heat is seen at $21.0 \text{ cm}^3/\text{mole}$ even at 2800 ppm ⁴He. Reich and Yu see results similar to those of Bernier and Landesman. Garwin and Reich see a factor of 4000 too much specific heat at $v = 19.3 \text{ cm}^3/\text{mole}$ for a ⁴He concentration of about 1%, 10000 ppm. The suggestion put forth here is an attempt at the explanation of this NMR specific-heat anomaly.

The basic idea of the explanation is that the excess specific heat in both the experiments mentioned above is due to the 4 He - 4 He interactions. We begin by arguing that even the specific-heat anomaly in the low-concentration-mixture data (e.g., that of Bernier and Landesman and Reich and Yu) is unusual. Mass fluctuation waves have been invoked to explain the relaxation mechanism-they do that pretty well-they will also explain the low-concentration specific-heat anomaly provided that the ⁴He particles tunnel through the ³He medium about ten times faster than ³He-³He tunneling rate. We take this need for a factor of 10 in J_{34} (i.e., $J_{34} \approx 10J_{33}$) as strong evidence that it is not the specific heat of the mass fluctuation waves that is being observed in these experiments.⁷

Assume the topology for relaxation in the experiments at Oxford, of Bernier and Landesman, of Reich and Yu, and of Garwin and Reich to be that shown in Fig. 1. The new *interaction* bath I appended to the ⁴He bath is due to the ⁴He-⁴He interactions.⁸ A long-range interaction occurs between two ⁴He atoms in a ³He medium because a ⁴He atom has less zero-point motion than a ³He atom and the ³He medium in the vicinity of a ⁴He atom expands toward the atom. A crude model for this process can be constructed using elastic continuum theory. A sphere embedded in a continuum gives rise to a



FIG. 1. Topology of the relaxation process. The kinetic and potential energy of the 4 He atoms is regarded as leading to two strongly coupled energy reservoirs.

radial pressure field about itself (either because it is larger or smaller than the piece of continuum it replaces). To place a second sphere at r away from the first one must do work against the pressure field to move the continuum to accomodate the second sphere.^{9,10} Thus there is an energy of interaction between the two spheres which is given approximately by

$$E(r) \approx -\epsilon^2 m_3 s^2 (\Delta/r)^3 , \qquad (1)$$

where s is the velocity of sound, Δ is the nearneighbor distance, and ϵ is a measure of the mismatch of the sphere. The volume difference is $4\pi\Delta^{3}\epsilon$. This interaction is long ranged. The *I* bath is taken to be due to this interaction, i.e.,

$$H_{I} = \sum_{RR'} E(|\vec{R} - \vec{R}'|) n_{4}(R) n_{4}(R') , \qquad (2)$$

where $n_4(R)$ is the number of ⁴He atoms at R. The manifold of states that constitute the I bath correspond to the various configurations of the ⁴He particles in the ³He medium. The energy for each configuration is given by Eq. (2). The coupling of the I bath to the tunneling bath comes about because the ⁴He particles move relative to one another by virtue of the tunneling process. As they do they exchange tunneling energy for interaction energy. Tunneling motion of a ⁴He particle is possible in regions of the sample where $|\nabla E_I(\vec{R}) \cdot \vec{\Delta}| < J_{34}$, where

$$E_{I}(\vec{R}) = \sum_{R' \neq (R)} E(|\vec{R} - \vec{R}'|) n_{4}(R') .$$
 (3)

In regions of the sample where this inequality is reversed, $|\nabla E_I(\vec{\mathbf{R}}) \cdot \vec{\Delta}| > J_{34}$, the uncertainty in energy of a ⁴He particle, an energy of about J_{34} , is not great enough to permit energy conservation to occur when the particles tunnel. Thus to move a ⁴He atom in a region of the sample of large $|\nabla E_r|$ requires assistance from the phonons or a higherorder process. But assistance from the phonons means that both tunneling energy and potential energy of the ⁴He atom are locked to the phonons. Thus we take the 4 bath and I bath of Fig. 1 to refer, respectively, to the tunneling energy and potential energy of the ⁴He particles⁸ in regions of the sample where $|\nabla E_I|$ is small, i.e., where $|\nabla E_I(\vec{R}) \cdot \vec{\Delta}| < J_{34}$. In these regions of the sample a single ⁴He particle moves among various con-

TABLE I. Values of J at selected molar volume. The calculations shown in Figs. 2 and 3 were done with the values of J listed here.

v (cm ³ /mole)	J (10 ⁻⁶ K)	ө _р (К)
19.3	5	38
20.0	23	29
21.0	60	26

figurations of the other ⁴He particles. It has energy of approximately β_T^{-1} to expend in visiting these various configurations; $\beta_T^{-1} = k_B T_T$, T_T is the temperature of the tunneling system. As the ⁴He particle moves in the low $|\nabla E_I|$ region of the sample it sees values of E_I that fluctuate about the average value \overline{E}_I . The important property of the various regions of the sample the ⁴He atoms visits is the fluctuation in E_I . These fluctuations come about because as a ⁴He moves through the low $|\nabla E_I|$ regions of the sample it sees fluctuations in the concentration of the other ⁴He atoms and corresponding fluctuations in E_I .

We begin by calculating the energy of a single ⁴He atom in the sample. We *assume* that the fluctuation in E_I attending a fluctuation Δx_4 in the concentration of ⁴He atoms is given by

$$\Delta E_I(\Delta x_4) = \lambda \epsilon^2 m s^2 \Delta x_4 , \qquad (4)$$

where λ is a constant of order 1. We may calculate the average energy of a ⁴He particle in the fluctuating field by the formula

$$\langle E_I(T) \rangle = \int d(\Delta x_4) P(\Delta x_4) \Delta E_I(\Delta x_4) e^{-\beta \Delta E_I(\Delta x_4)} ,$$
(5)

where $P(\Delta x_4)$ is the probability of a fluctuation in concentration of magnitude Δx_4 . Since

$$\int d(\Delta x_4) P(\Delta x_4) = 1, \quad \int d(\Delta x_4) \Delta x_4 P(\Delta x_4) = 0,$$

in the high-temperature limit $\beta \Delta E_I(\Delta x_4) \ll 1$ we have

$$\langle E_I(T) \rangle \approx \lambda^2 \left[(\epsilon^2 m s^2)^2 / k_B T \right] \langle \Delta x_4^2 \rangle$$
 (6)

But

$$\langle \Delta x_4^2 \rangle = \int d(\Delta x_4) \, \Delta x_4^2 \, P(\Delta x_4) = x_4 \tag{7}$$

so that Eq. (6) yields

$$\langle E_I(T) \rangle \approx \lambda^2 \left[(\epsilon^2 m s^2)^2 / k_B T \right] x_4$$
 (8)

This is the average interaction energy of a single ⁴He atom. We take the total number of ⁴He atoms in low $|\nabla E_I|$ regions of the sample to be proportional to the total number of ⁴He atoms ANx_4 , where A is a proportionality constant. Thus the average interaction energy of the I bath is

$$\langle E_I(T) \rangle / N = C - A \lambda^2 [(\epsilon^2 m s^2)^2 / k_B T] x_4^2;$$
 (9)

here C is a temperature-independent constant representing the temperature-independent interaction energy of the ⁴He atoms. The specific heat of the I bath is

$$C_I(T)/Nk_B = A \lambda^2 x_4^2 (\epsilon^2 m s^2/k_B T)^2$$
 (10)

For the relaxation time in the topology shown in Fig. 1 we have⁹

$$\frac{1}{T_1} = \frac{1}{T_{2\,\rho}} \, \frac{k_4}{k_T + k_Z + k_I + k_4} \quad , \tag{11}$$

where T_{2p} is the mass fluctuation phonon-relaxation time (two phonon, 2p), $k_T \propto J^2$, $k_4 = Bx_4k_T$, $k_Z \propto \omega_0^2 \ll J^2$ in low Larmor field, $B = (J_{34}/J)^2 \approx 1$, and $k_I = A\lambda^2 (\epsilon^2 m s^2)^2 x_4^2$. Thus

$$\frac{1}{T_1} = \frac{1}{T_{2p}} \frac{x_4}{1 + x_4 + (k_I / k_T)} \quad . \tag{12}$$

Now for $\epsilon \approx 10^{-2}$ we have $\epsilon^2 m s^2 \approx 0.01$ K and

$$k_I / k_T \approx A \lambda^2 x_4^2 (0.01/J)^2$$
 (13)

At $v = 20.0 \text{ cm}^3/\text{mole}$, $J \approx 20 \ \mu\text{K}$, k_I/k_T is order 1 at $A\lambda^2 x_4^2 = 4 \times 10^{-6}$. We may use the results of Bernier and Landesman and assume the value 1 obtains for $x_4 = 1000 \text{ ppm}$. Then, we have $A\lambda^2 \approx 1$. We will use this value of $A\lambda^2$ for all molar volumes¹¹ (19.3, 20.0, and 21.0) for which we discuss T_1 . We take T_1 to be given by

$$\frac{1}{T_1} = \frac{1}{T_{2p}} \frac{x_4}{1 + 10^{-4} (x_4^2/J^2)} \left(\frac{J_{34}}{J}\right)^2 .$$
(14)

In Table I, we have tabulated the parameters that characterize solid 3 He at molar volumes of 19.3, 20.0, and 21.0. Using these parameters we have



FIG. 2. Concentration dependence of T_1 . The inverse of the topological factor in Eq. (14) yields the concentration dependence of T_1 and is plotted here as a function of concentration for three molar volumes. Operating points for the relevant experiments are shown.



FIG. 3. Excess specific heat. The excess specific heat k_T/k_T from Eq. (13) is plotted as a function of concentration. Operating points for the relevant experiments are shown.

evaluated the topological factor in Eq. (14), i.e., the factor $x_4/[1+10^{-4}(x_4^2/J^2)]$. The inverse of this factor is proportional to T_1 and is plotted in Fig. 2. From Fig. 2 we see the concentration dependence of T_1 ; T_1 decreases with x_4 at small values of x_4 and then increases for values of x_4 beyond about 10^{-3} or 10^{-4} . On Fig. 2 we have indicated the concentrations at which the T_1 experiments of the Oxford group and Garwin and Reich have been done. We see that due to the topological factor alone the value of T_1 in these two experiments should differ by a factor of about 100. Further at v = 19.3 the two-phonon process (it goes as Θ_D^{-6}) is about a fac-

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⁷Actually the explanation of T_1 in terms of the mass fluctuation waves, although quantitatively correct, pushes

tor of 5 slower than it is at v = 20.0. Thus from Eq. (14) we are led to expect T_1 seen by Garwin and Reich $(x_4 = 1\%)$ to be about 500 times longer than the T_1 seen by Giffard and Hatton ($x_4 = 300$ ppm) at T = 0.4 K. At T = 0.4 K, $x_4 = 0.01$, Garwin and Reich see $T_1 \approx 6000$ sec; at T = 0.4 K, $x_4 = 300$ ppm, Giffard and Hatton see $T_1 \approx 3$ sec. The ratio of these two experimental values of T_1 , 2000, is within a factor of 4 of the result predicted by Eq. (14). Further the factor $(J_{34}/J)^2$ in Eq. (14) that we have taken to be 1 for both v = 19.3 and v = 20.0works in the right direction to bring better agreement between Eq. (14) and experiment. We believe Eq. (14) correctly explains the qualitative features of the concentration dependence of T_1 as seen by Garwin and Reich and Giffard and Hatton.

From Eq. (14) we see that at v = 19.3 and $x_4 = 0.01$ the denominator of the topological factor is 3500, i.e., there is at least three orders of magnitude more specific heat in the ${}^{4}\text{He} - {}^{4}\text{He}$ interaction than there is in the tunneling motion of the entire ³He bath. We believe Eq. (14) correctly explains the anomalous specific heat seen by Garwin and Reich, Fig. 3.

Equation (14) does not agree with the linear excess specific heat at low concentrations seen by Bernier and Landesman and Reich and Yu. However, it does say that much less excess specific heat will be seen at $v \approx 21.0$ than at v = 20.0 at a given concentration. This latter result agrees with the findings of Bernier and Landesman and Reich and Yu.

The excess specific heats we are discussing here are in all cases at least four orders of magnitude smaller than the phonon specific heat. These specific heats cannot be seen in conventional thermostatic measurements-they are seen in NMR experiments because the phonons are out of the picture on a time scale of order T_1 .

¹⁰R. A. Guyer (unpublished).

¹¹We emphasize that this procedure for determining $A\lambda^2$ is highly conditional on the estimate of the strength $\epsilon^2 m s^2$. In fact the experiment of Bernier and Landesman can only fix the product $A\lambda^2 (\epsilon^2 m s^2)^2$.

¹R. L. Garwin and H. A. Reich, Phys. Rev. Letters 12, 354 (1964). ²M. Bernier and A. Landesman, Solid State Commun.

^{7, 529 (1969).} ³H. A. Reich and W. N. Yu, Solid State Commun. <u>7</u>,

^{1521 (1969).}

⁴R. A. Guyer and L. I. Zane, Phys. Rev. Letters <u>24</u>, 660 (1970).

⁵R. P. Giffard and J. Hatton, Phys. Rev. Letters <u>18</u>, 1106 (1967).

⁶R. P. Giffard, thesis (Oxford University, 1968) (unpublished).

the physical picture of a separation of the motion of the ⁴He atoms from the motion of the ³He medium too hard. A less extreme view is that put forth by Tsuneto and Yamashita (unpublished).

⁸Here with regard to the role of the ⁴He-⁴He interaction we are once again taking an overly simplified view (see Ref. 7). Our justification is that we are attempting to identify the essential physical process.

⁹R. A. Guyer, R. C. Richardson, and L. I. Zane, Rev. Mod. Phys. <u>43</u>, 532 (1971).