

while<sup>6</sup>

$$-\left(\frac{d\mu}{dT}\right)_v = \frac{\pi^2}{3} k^2 T \left(\frac{d \ln D}{d\epsilon}\right)_{\epsilon=\mu}, \quad (3)$$

with  $\sigma$  the conductivity and  $D$  the energy density of states. While the electrons in Ni are by no means independent and the finite compressibility near  $T_C$

should not be ignored, the similarity of the right-hand sides of Eqs. (2) and (3) does tempt one to speculate that the changes in transport properties near  $T_C$  are due principally to changes in the density of electronic states.

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<sup>2</sup>D. L. Connelly, J. S. Loomis, and D. E. Mapother, Phys. Rev. B **3**, 924 (1971).

<sup>3</sup>C. A. Domenicali, Rev. Mod. Phys. **26**, 237 (1954).

<sup>4</sup>This point is clearly explained by J. M. Ziman [in *Electrons and Phonons* (Oxford U.P., London, 1960), p. 383].

<sup>5</sup>N. F. Mott and H. Jones, *Theory of the Properties of Metals and Alloys* (Oxford U.P., London, 1936), p. 178.

<sup>6</sup>Reference 5, p. 310.

## Reply to "Comment on 'Seebeck Coefficient at the Curie Point Temperature: Specific Heat of Charge Carrier in Ferromagnets'"

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### INTRODUCTION

Recently, we have attempted a more complete analysis of the critical behavior of the Seebeck coefficient in conducting ferromagnets<sup>1</sup> than was possible in our letter on Ni.<sup>2</sup> Although we have discussed some of the points raised by Taylor in the previous comment<sup>3</sup> in this recent work, we will clarify some of the Taylor's specific points here.

First, contrary to Taylor's introductory remarks, it is now quite generally accepted that it is precisely the same fluctuations which govern the static properties near a critical point that also govern the most accessible of the transport properties, the resistivity.<sup>4,5</sup> The temperature coefficient of the resistivity has been shown to be closely related to the specific heat in a number of systems.<sup>6,7</sup>

Second, nowhere in our paper did we relate the chemical potential  $\mu$  with the "transport entropy"; our Eq. (1) relates the electrochemical potential  $\bar{\mu}$  to the transport entropy through a Gibbs-Duhem

relation, and this is *not* equivalent to Taylor's Eq. (1). Indeed, the chemical potential is not related to the Seebeck coefficient by any thermodynamic argument there; it was the purpose of our paper to indicate the strong empirical correlation between the critical component only of these two quantities, and then only when the  $vdp$  term is small as is the case for Ni. For other systems, such as Gd along the  $c$  axis, the  $vdp$  term can dominate the behavior.<sup>8</sup>

Finally, we agree with Taylor that the Seebeck coefficient and the density of states are closely related, but this point is not new. The argument was put forward by Mott<sup>9</sup> and is referenced four pages after Taylor's Ref. 6. Unfortunately, the calculation only attempts to explain the critical behavior for temperatures below the transition.

In closing, we remark that our new analysis<sup>1</sup> strongly supports the hypothesis of our letter on Ni. The thermoelectric effect can be used to analyze the contribution of the charge carriers to

the critical effects associated with ordering, be it order-disorder, localized-moment ferromagnetic, or itinerant-electron ferromagnetic. More

systematic studies of the Seebeck coefficient and exploratory work on the other transport properties are now needed.

<sup>1</sup>S. H. Tang, T. A. Kitchens, F. J. Cadieu, and P. P. Craig, in Proceedings of the International Conference on Low-Temperature Physics, Boulder, Colorado, August, 1972 (unpublished).

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<sup>3</sup>P. L. Taylor, preceding paper, Phys. Rev. B **7**, 1197 (1973).

<sup>4</sup>P. P. Craig, W. I. Goldburg, T. A. Kitchens, and J. I. Budnick, Phys. Rev. Lett. **19**, 1334 (1967); P. C. Zumsteg and R. D. Parks, Phys. Rev. Lett. **24**, 520 (1970).

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<sup>6</sup>D. S. Simons and M. B. Salamon, Phys. Rev. Lett. **26**, 750 (1971).

<sup>7</sup>An excellent review of other resistivity studies is given by R. D. Parks [*Magnetism and Magnetic Materials* (AIP, New York, 1971), p. 630].

<sup>8</sup>F. J. Cadieu and D. H. Douglass, Phys. Rev. Lett. **21**, 680 (1968).

<sup>9</sup>N. F. Mott, Proc. R. Soc. A **156**, 368 (1936).

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## Nuclear Magnetic Resonance in NbAl Alloys

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Measurements of the Knight shift of Nb<sup>93</sup> were made for Nb-Al alloys in the body-centered-cubic solid-solution region up to 8-at.% Al in Nb and in the two-phase region at 14-at.% Al. In the solid solution, the Knight shift decreases from 0.845% in pure Nb to 0.780% at 8-at.% Al. In the two-phase region made up of Nb and Nb<sub>3</sub>Al, the Knight shift is 0.805%. The decrease in the shift vs Al content is tentatively interpreted according to the rigid-band model. This is in contrast to previous work in V-Al and V-Ga systems where the rigid-band model is not believed to apply.

The <sup>93</sup>Nb Knight shift has been measured in Nb-Al alloys in the bcc solid-solution region and in the two-phase region consisting of Nb and the intermetallic compound Nb<sub>3</sub>Al. Previous studies on alloys of transition metals with other transition metal<sup>1</sup> have been interpreted according to the rigid-band model. In this model, the density of electronic states is based on the host transition metal. The alloying elements with greater or smaller valence than the host serve to increase or decrease the Fermi energy, respectively, leaving the shape of the host-band unaltered. In fact, the increase or decrease does not depend on the solute but only on the number of electrons per atom  $\delta$ . Thus, for example, Cr or Fe alloyed with V have essentially the same effect on the electronic properties of V when their concentration have the same  $\delta$ .

According to this model, the Knight shift and magnetic susceptibility are partitioned into portions associated with *s*, *d* electrons, and the orbital paramagnetism. The partitioning is performed through a knowledge of the electronic density of states obtained through the use of low-temperature specific-

heat data.

However, when Al or Ga is alloyed with V, the rigid-band model can no longer be applied to interpret the results. For these alloys the Al or Ga conduction electrons seem to be independent of the V *d* band. It is as though the wave functions of the V *d* band electrons avoid the Al cells, as if the Al were an obstacle. The current results on Nb-Al alloys appear to be consistent with the rigid-band model, perhaps indicating that the classification into transition-transition and transition-non-transition alloys is not entirely correct.

The alloys were prepared by arc melting the required amounts of zone-refined Nb and 99.999-wt.% Al in an argon atmosphere. Since the weight of the samples varied less than 0.01% for a 1-g sample after melting, the alloy composition reported here is that of the weighed charges before melting. Each specimen was homogenized at 1050 °C for one week and then heat treated for one week at 900 °C. All heat treatments were in a vacuum and the specimens were subsequently water quenched.

The solid solubility of aluminum in niobium de-