Comment on the isotope effect in weak itinerant ferromagnets

J. Appel
I. Institut für Theoretische Physik, Universität Hamburg, Hamburg, West Germany

D. Fay
Abteilung für Theoretische Festkörperphysik, Universität Hamburg, Hamburg, West Germany

(Received 29 January 1980)

The isotope effect on the Curie temperature is evaluated using our recent disscussion of the phonon contribution to the Stoner enhancement factor. An isotope effect exists only if the phonon contribution to the short-range Hubbard-type exchange interaction contains the small Migdal factor. The value of $\alpha = (M/T_C)(dT_C/dM)$ is ≈ 1 or $\ll 1$ depending on whether or not the phonon contribution is pertinent for the magnetic transition.

The electron-phonon contribution to the Stoner enhancement of the Pauli spin susceptibility is of some interest for the magnetic properties of $ZrZn_2$ and of other metallic substances that become weak, itinerant ferromagnets at low temperatures.[1-3] The phonon contribution to the short-range Hubbard-type exchange interaction should lead to an observable effect, namely the isotope effect on the Curie temperature. This effect was first predicted by Hopfield[4(a)] in a qualitative manner. It is the purpose of this note to evaluate this effect based on our recent discussion of the phonon contribution to the Stoner factor S.[3]

By definition, for finite temperature T,

$$S(T) = \lim_{\vec{q}\to 0} \lim_{q_0 \to 0} \frac{\chi(q,T)}{\chi_0(q,T)} = \frac{1}{1 - I\chi_0(0,T)} \quad , \tag{1}$$

where χ and χ_0 are the susceptibilities of the interacting and noninteracting systems, respectively, and $q = (\vec{q}, q_0)$. By taking Eq. (1) as our starting point we ignore Moriya's spin fluctuation part in the free energy and hence in χ that is caused by mode-mode coupling and that has been calculated by Gumbs and Griffin.[4(b)] The corresponding effect on α is small. The irreducible particle-hole interaction I is assumed to be temperature independent and consists of the Hubbard exchange interaction (equal to the intra-atomic Coulomb repulsion) I_c and the phonon contribution $I_{\text{el-ph}}$. For convenience, we redefine the quantity $I\chi_0(0,T)$ by writing

$$I\chi_0(0,T) = \bar{I}(T) = I_c \chi_{\text{phon}}(0,T) \quad , \tag{2}$$

where χ_{phon} is the bubble diagram χ_0 completely renormalized by phonons, that is, by the zero-point vibrations of the lattice. The quantity χ_{phon} is discussed in Ref. 3. The Hartree-Fock criterion for ferromagnetism is that S becomes infinite. Under this condition the Curie temperature T_C is given by

$$\bar{I}(T_C) \approx I_c \chi_{\text{phon}}(0,0)\left\{1 + \tfrac{1}{6}\pi^2 \frac{N''(E_F)}{N(E_F)}(k_B T_C)^2\right\} = 1 \quad , \tag{3}$$

where the $\approx$ indicates that the temperature dependence of $\chi_{\text{phon}}(0,T)$ is approximately given by that of $\chi_0(0,T)$ for $k_B T_C \ll E_F$ (equal to Fermi energy at temperature T), that is, for weak itinerant ferromagnets. Here $N(E_F)$ is the bare density of states per spin at E_F.[4(c)] For a parabolic band, we have

$$N''(E_F)/N(E_F) = -1/4E_F^2 \quad .$$

From Eq. (3) we get

$$k_B T_C = \left| \frac{6}{\pi^2} \frac{N(E_F)}{N''(E_F)} \frac{1-\bar{I}}{\bar{I}} \right|^{1/2} \quad , \tag{4}$$

where $\bar{I} = I_c \chi_{\text{phon}}(0,0)$. Equation (4) yields a finite value of T_C if $\bar{I} > 1$ and $N''(E_F) < 0$. This case applies not only to a parabolic band but also to the case where E_F lies near a peak in the density of states as indicated for $ZrZn_2$ by recent band-structure calculations (cf. Fig. 1).[5-9]

The isotope effect on the T_C is given by

$$\alpha = \frac{M}{T_C}\frac{dT_C}{dM} = \frac{1}{2}\frac{I_c}{\bar{I}(\bar{I}-1)} M \frac{d\chi_{\text{phon}}}{dM} \quad , \tag{5}$$

where M is the ionic mass and $\chi_{\text{phon}} \equiv \chi_{\text{phon}}(0,0)$. Since it is a difficult task to evaluate χ_{phon} to order ω_D/E_F (equal to the ratio of the Debye energy to the Fermi energy), we presented in Ref. 3 a qualitative discussion of the three different bubble diagrams that involve a single phonon line only. In particular we estimated the contribution of the "phonon-exchange" bubble [cf. Fig. 3(a) of Ref. 3] from an analysis of the lowest-order phonon correction to the electron-phonon vertex. The result can be written in

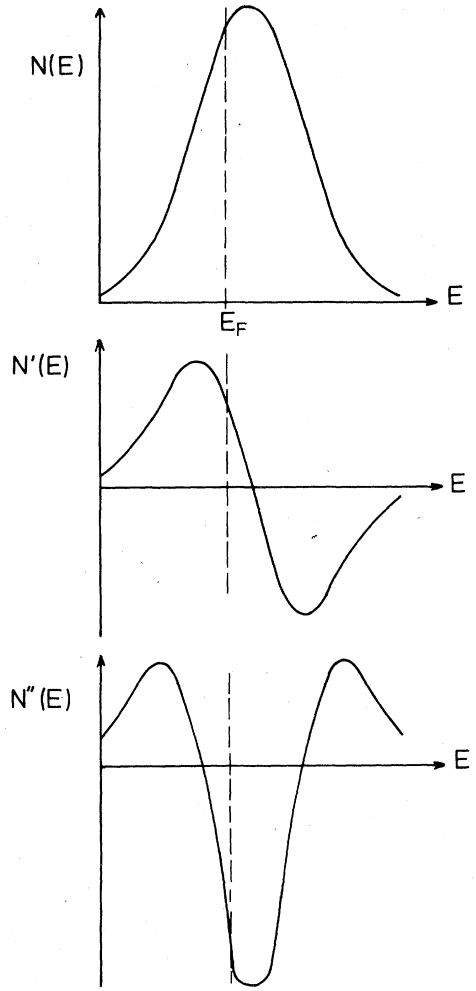

FIG. 1. Density-of-states peak, $N(E)$, and its first two derivatives near the Fermi level E_F (schematic); E_F falls on the "low side" of the peak as in Refs. 6 and 7.

the form

$$I_c \chi_{\text{phon}} = \bar{I}_c + \bar{I}_{\text{el-ph}} , \tag{6}$$

$$\bar{I}_c = I_c N(E_F) , \quad \bar{I}_{\text{el-ph}} = \epsilon(\omega_D/E_F)\lambda_{\text{BCS}} ,$$

where λ_{BCS} is the BCS coupling constant and ϵ is a parameter of order one, the sign of which we have not been able to determine. We point out that the entire M dependence of χ_{phon} is due to the "Migdal factor" (ω_D/E_F) in $\bar{I}_{\text{el-ph}}$. If $\bar{I}_{\text{el-ph}}$ were simply equal to λ_{BCS}, as suggested in Refs. 1 and 2, then there should be no isotope effect at all. Substitution of Eq. (6) into (5) gives

$$\alpha = -\frac{1}{4}\frac{\bar{I}_{\text{el-ph}}}{\bar{I}(\bar{I}-1)} , \tag{7}$$

with $\bar{I}_{\text{el-ph}}$ given below Eq. (6). In order to estimate α let us assume that $\bar{I}_c = 1$, i.e., that the ferromagnetism corresponding to $\bar{I} > 1$ is due to the small phonon contribution $\bar{I}_{\text{el-ph}}$. It is immediately seen from Eq. (7) that, in this special case, $\alpha \cong -\frac{1}{4}$. A value of $|\alpha| \approx 1$ would seem to be a reasonable upper limit for the magnitude of this effect.

We now wish to discuss the experimental attempt to measure α in $ZrZn_2$ by Knapp, Corenzwit, and Chu[10] in terms of Eq. (7). The authors find that the best experimental value to describe the Zr-isotope effect on T_C is given by

$$\alpha_{Zr} = 0.2 \pm 0.2 . \tag{8}$$

This value of α_{Zr} is arrived at after the effect of impurity scattering on the T_C's of the four different Zr isotopes is corrected for by assuming a linear relationship between T_C and T_s, the superconducting transition temperature of Zr metal which exhibits no isotope effect. The agreement between Eqs. (7) and (8), assuming that $\bar{I}_{\text{el-ph}} = \bar{I} - 1$, that is, the electron-phonon interaction tips the balance towards ferromagnetism, may be accidental but it appears that the large isotope effect predicted before[2,4(a)] is in conflict with Migdal's theorem. This conclusion gets additional support when we calculate α using recent band-structure calculations.[5-9] All of the authors seem to agree that the Fermi level E_F is situated in a huge peak in the total density of states as indicated in Fig. 1, and that E_F falls on the "low side" of the peak (cf., e.g., Ref. 6). Because of this location where $N(E)$ changes rapidly, the calculated values of $N(E_F)$ differ somewhat, the numerical values range from 42 states per Ry spin $ZrZn_2$ in Ref. 7 to 64 in Ref. 5. Glötzel and Andersen[7] also calculate the intra-atomic Coulomb interaction at a Zr atom and find $I_c = 42$ mRy, hence they quote $\bar{I}_c = 1.17$. Taking their numbers at face value and assuming thereby that the balance towards magnetism in $ZrZn_2$ is not significantly affected by phonon "exchange" we get, setting $\bar{I} = \bar{I}_c$ in Eq. (7), $\alpha = -1.3\bar{I}_{\text{el-ph}}$. To estimate $\bar{I}_{\text{el-ph}}$ we take the Debye temperature, $\Theta_D = 370\,^\circ\text{K}$.[11] For the Fermi energy E_F we take the value, 0.075 Ry, given by the band structure[7] when the Fermi level is measured whith respect to the $\Gamma_{25'}$ level. As pointed out in Ref. 7, the energy band responsible for the peak in the density of states near E_F is the one going from the $\Gamma_{25'}$ level to the uppermost X_4 and $L_{3'}$ levels (cf. Fig. 2 of Ref. 7). The above value for E_F should be a fair estimate for the $4d$ electrons at the Fermi surface to delocalize themselves, relevant for Migdal's theorem. Using these values, $\omega_D/E_F = 0.03$, we get $\alpha = -0.04\epsilon\lambda_{\text{BCS}}$. This α value is much smaller than the one calculated above under the assumption $\bar{I}_{\text{el-ph}} = \bar{I} - 1$. However, this α value, too, is within the experimental bounds given by Eq.(8).

Hence, the present experimental state of affairs for the isotope effect on T_C in $ZrZn_2$ does not allow for a conclusion on whether or not $\bar{I}_{\text{el-ph}}$ is important in that it tips the balance towards the observed ferromagnetism. Equation (8) does, however, indicate that $\bar{I}_{\text{el-ph}}$ is probably positive. If the small phonon effect plays a pertinent role, $\bar{I}_{\text{el-ph}} \cong I - 1$, we expect the isotope effect to be large, that is $|\alpha| \lesssim 1$. If, on the other hand, the weak ferromagnetism is caused primarily by the ordinary exchange interaction between electrons, we expect $|\alpha| << 1$. An experimental attempt to measure α with a small error margin on clean samples whould help to determine whether or not phonons play a significant role for T_C.

Note added in proof. We take λ_{BCS} as independent of M and attribute the deviation of the *superconducting* isotope effect from its normal value of $-1/2 (T_{\text{superc.}} \propto M^{-1/2})$ as being entirely due to the Coulomb pseudopotential for the scattering between electrons near the Fermi surface [see J. R. Schrieffer, *Theory of Superconductivity* (Benjamin, New York, 1964), p. 187.]

[1]C. P. Enz and B. T. Matthias, Z. Phys. B 33, 129 (1979).
[2]C. P. Enz, in *Superconductivity in d- and f-band metals*, edited by H. Suhl and B. Maple (Academic, San Francisco, 1979).
[3]D. Fay and J. Appel, Phys. Rev. B 20, 3705 (1979).
[4](a) J. J. Hopfield, Phys. Lett. A 27, 397 (1968); (b) G. Gumbs and A. Griffin, Phys. Rev. B 13, 5054 (1976); (c) In terms of $E_F^0 = E_F(T=0)$, we have $N''(E_F)/N(E_F) = \{N''(E_F^0)/N(E_F^0) - [N'(E_F^0)/N(E_F^0)]^2\}$.
[5]T. Jarlborg and A. J. Freeman (unpublished).
[6]R. A. de Groot, D. D. Koelling, F. M. Mueller, B. T. Matthias, A. L. Giorgi, G. T. Stewart, and J. L. Smith (unpublished).
[7]D. Glötzel and O. K. Andersen (unpublished).
[8]D. D. Koelling, D. L. Johnson, S. Kirkpatrick, and F. M. Mueller, Solid State Commun. 9, 2039 (1971).
[9]D. L. Johnson, Phys. Rev. B 9, 2273 (1974).
[10]G. S. Knapp, E. Corenzwit, and C. W. Chu, Solid State Commun. 8, 639 (1970).
[11]F. E. Hoare and J. C. G. Wheeler, Phys. Lett. 7, 402 (1966).