ing. When the fluorescence line moves to a position more or less symmetrical with respect to two cavity modes, both could oscillate. This is believed to be responsible for some of the 230-Mc/sec beat signals observed in our experiment, especially those appearing not at the onset of the laser pulse. This can obviously be avoided by using a shorter optical feedback path. The residual pulling effect can again be compensated in the same way as that for the cavity shift.

At low temperatures ($\lesssim 77°K$), the fluorescence line shape becomes more complicated due to the fact that the $^4A_2$ ground state of Cr^{3+} in Al_2O_3 is actually split by about 0.38 cm^{-1} due to spin-orbit interaction in a crystal field of C_3 symmetry.[12] In addition, as the temperature is reduced, although the total fluorescence linewidth is reduced, the ratio of the temperature-dependent part of the width to the temperature-independent strain broadened width decreases. When the separation between the cavity resonances is larger than the temperature-dependent part but still smaller than the strain broadened part of the total linewidth, one would then expect that the oscillating modes will be able to burn holes into the fluorescence line, and more than one mode will again be able to oscillate as the two-phonon Raman process becomes less and less effective in quenching other oscillating modes. Preliminary experimental results indicate this to be the case, although we have not yet obtained conclusive results due to the difficulties involved in keeping the laser rod at the desired low temperature.

[12] J. E. Geusic, Phys. Rev. **102**, 1252 (1956); A. L. Schawlow, *Advances in Quantum Electronics*, edited by J. R. Singer (Columbia University Press, New York, 1961), p. 232.

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Strong Phonon Effects in High-Transition-Temperature Superconductors

A. M. CLOGSTON
Bell Telephone Laboratories, Murray Hill, New Jersey
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The high-transition-temperature superconducting compounds V_3Ga and V_3Si have a very large electronic specific heat and strong temperature-dependent susceptibility. These properties are interpreted in terms of a large electron-phonon enhancement of the density of states, and a large exchange enhancement of the susceptibility.

WE have reported previously on some of the unusual properties of the high-temperature superconductors crystallizing in the β-wolfram system.[1,2] These unusual properties include a strongly temperature-dependent susceptibility and Knight shift,[1,2] and a very large low-temperature electronic specific heat.[3] In our original interpretation of these properties, we suggested that the band structure of V_3Ga and V_3Si is such that a high, narrow peak in density of states exists at the Fermi level. We now wish to modify this interpretation. We shall advance a hypothesis below that leads to the conclusion that the susceptibility and specific heat of V_3Ga and V_3Si are strongly affected, respectively, by Coulomb exchange interactions and electron-phonon interactions. The effect of these interactions is to exaggerate the temperature dependence of the susceptibility and to greatly increase the electronic specific heat at low temperatures. In Ref. 2, the temperature-dependent Knight shift has been used to identify a temperature-independent orbital contribution to the sussceptibility of V_3Ga and V_3Si. Various uncertainties place the orbital susceptibility in the range $5.2-7.5\times10^{-4}$ emu/mole. To simplify the considerations that follow we shall adopt the value 7.0×10^{-4} emu/mole as being close to the correct value. The value chosen will not affect the qualitative aspects of the arguments to be made. We shall take χ_{mole} to be the susceptibility of V_3Ga or V_3Si in units of emu per mole due to the spin paramagnetism, and equal to the measured susceptibility minus the orbital susceptibility. We neglect the small Landau diamagnetism. If χ_a is the susceptibility per atom corresponding to χ_{mole}, $\chi_a/2\mu_B^2$ has units of states per erg per atom. In more convenient units we have $\chi_a/2\mu_B^2$ (states per electron volt per atom)$=1.55\times10^4\ \chi_{\text{mole}}$. In Fig. 1 we have plotted $\chi_a/2\mu_B^2$ as a function of temperature for V_3Ga[1] using units of states per electron volt per vanadium atom. We also indicate the density of states at the Fermi level $\eta(E_f)=7.1$ states/eV-atom derived from the low-temperature specific heat.[3]

[1] A. M. Clogston, and V. Jaccarino, Phys. Rev. **121**, 1357 (1961).
[2] A. M. Clogston, A. C. Gossard, V. Jaccarino, and Y. Yafet, Phys. Rev. Letters **9**, 262 (1962).
[3] F. J. Morin and J. P. Maita, Phys. Rev. **129**, 1115 (1963).

From perturbation theory, or by use of the random-phase approximation,[4] it can be shown that the spin susceptibility and density of states may be written as

$$\chi = 2\mu_B^2 \eta_0(E_f)/[1-\eta_0(E_f)J], \quad (1)$$

$$\eta(E_f) = \eta_0(E_f)/[1-\eta_0(E_f)S], \quad (2)$$

where J arises solely from the Coulomb interactions, and S arises largely from the lattice interactions in the range of metallic densities. The density of states for the noninteracting gas is $\eta_0(E_f)$. Equations (1) and (2) can be combined to give

$$\chi = 2\mu_B^2 \eta(E_f)/[1+\eta(E_f)(S-J)]. \quad (3)$$

Thus, χ and η are both increased over the values expected for a noninteracting gas, and χ may be larger or smaller than the value expected from the measured density of states depending on whether S is larger or smaller than J. If the dielectric constant of the electron gas is approximated by $E(q)=1+\sigma^2/q^2$, we have[4]

$$J = (2\pi e^2/\Omega k_f^2)\{1-(\sigma/2k_f)^2 \ln[1+(2k_f/\sigma)^2]\}, \quad (4)$$

where k_f is the Fermi momentum, Ω is the atomic volume, and the shielding constant σ is given by

$$\sigma^2 = 8\pi e^2 \eta_0(E_f)/\Omega. \quad (5)$$

We introduce a quantity g of dimensions of energy by the relation

$$g = \tfrac{2}{3}(e^2/a_0)(a_0/r)(3/2\pi N)^{2/3}, \quad (6)$$

where r is the radius of a sphere of volume Ω, N is the number of valence electrons per atom, and $a_0=\hbar^2/me^2$. Equation (4) can then be written

$$J = g[1-\eta_0 g \ln(1+1/\eta_0 g)]. \quad (7)$$

We now adopt a two-part hypothesis: (a) that Eqs. (1) and (2) hold for V_3Ga; and (b) that we can make a reasonable estimate of J from Eq. (7). From (a) we argue as follows: The value of $\chi_a/2\mu_B^2$ extrapolated to $T=0$ is 5.6 states/eV-atom, while η has the value 7.1 states/eV-atom. From Eq. (3) we can then obtain $(S-J)=0.038$ eV. Because of the uucertainties in the orbital susceptibility, we cannot claim high accuracy for this value. The corresponding value of η $(S-J)$ is 0.27, and we have previously pointed out that this is consistent with the high-superconducting transition temperature of V_3Ga.[2]

From (b) we proceed to estimate J by setting $N=5$ for the vanadium atoms in V_3Ga and choosing $2r$ equal to the distance between nearest-neighbor atoms so that $r/a_0=2.28$. We then obtain $g=1.66$ eV. We tentatively take $\eta_0=3.1$ states/eV-atom (compare below) and arrive at the estimate $J=0.143$ eV. Combining this with the result above, we have $S=J+0.038=0.181$ eV. Then, from Eq. (2), $\eta_0=3.1$ states/eV-atom. We are led to the conclusion that the density of states in V_3Ga has been enhanced by phonon effects by a factor $7.1/3.1=2.3$.

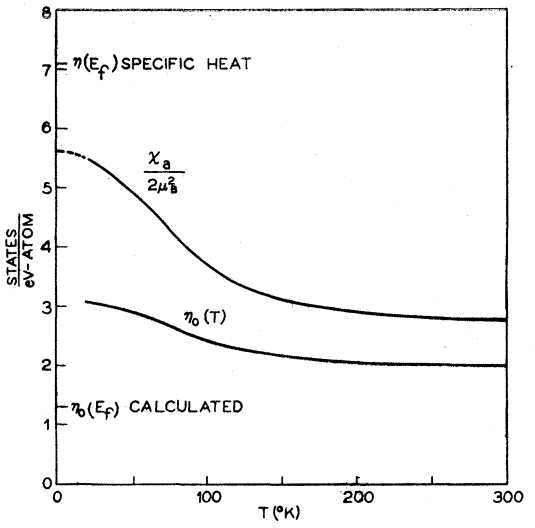

FIG. 1. Susceptibility and unenhanced density of states as a function of temperature for V_3Ga. Values are also shown for the density of states determined by the low-temperature specific heat, and for the density of states calculated by the augmented plane-wave method.

At finite temperatures, the spin susceptibility should be written as

$$\chi(T) = 2\mu_B^2 \eta_0(T)/[1-\eta_0(T)J], \quad (8)$$

where by $\eta_0(T)$ we mean

$$\eta_0(T) = \int \eta_0(E)(-)f'(E)dE, \quad (9)$$

with $f(E)=1/[\exp(E-E_f/kT)+1]$. We can calculate $\eta_0(T)$ from the experimental values of $\chi(T)$ and the value of J obtained above. The result is shown in Fig. 1. The thermal averaging over density of states evident in $\eta_0(T)$ is much less marked than in $\chi(T)$, where the effect is exaggerated by exchange. We consequently require a much less extreme variation of $\eta_0(E)$ with energy in order to explain the temperature dependence of $\eta_0(T)$. The thermal average of the density of states near the Fermi level at 300°K is approximately 2.0 states/eV-atom.

The hypothesis adopted above thus accounts for the unusual properties of V_3Ga in terms of a density of states that changes from a value of 3.1 states/eV-atom at the Fermi level to a value somewhat less than 2.0 states/eV-atom within an energy range of about 150°K. This is a more physically reasonable picture than the previous interpretation which would require the density of states to vary from about 5.6 to less than 2.8 states/eV-atom in the same energy range.

A recent calculation of the band structure of the β-wolfram compounds by the augmented plane-wave

[4] A. M. Clogston, V. Jaccarino, and Y. Yafet, Phys. Rev. 134, A650 (1964).

method has been carried out by Mattheiss.[5] For V_3Ga, he finds the Fermi level situated near a peak in the density of states of height 1.3 states/eV-atom and width 0.27 eV (3100°K). The calculation is carried out to an accuracy of about 0.15 eV with the result that structure in the peak is not resolved. We may therefore compare the calculated value of 1.3 states/eV-atom with the thermally averaged value of $\eta_0=2.0$ states/eV-atom. In view of the uncertainties of the two ways of determining η_0, this agreement may be considered good. Suppose, however, that no electron-phonon effects were present. One would then again require an extremely high, narrow peak in the density of states in order to reconcile the experimental low-temperature value of $\eta=7.1$ states/eV-atom with the calculated value of 1.3 states/eV-atom. It does not appear likely that any improvement in accuracy of the calculation would reveal such a peak and coincidently place the Fermi level exactly at the peak.

This note is written to call attention to the possibility of very large phonon effects in the specific heat of V_3Ga and V_3Si. If such large effects exist in these intermetallic compounds, important effects probably exist also in the transition metals, particularly in metals and alloys with about 3 and 4.5 valence electrons per atom, where the observed density of states is large. In the case of vanadium, the observed specific heat is 2.21×10^{-3} cal/°C-mole corresponding to a density of states of 1.96 states/eV-atom. A recent discussion by Krebs[6] suggests that this value has been enhanced by electron-phonon interactions by a factor of 1.9. We may also compare with a value for $\eta_0=1.25$ states/eV-atom taken from a calculation by Wood[7] for bcc iron.

A particularly interesting case is presented by scandium. The available information for this metal has been summarized in a recent paper by Gardner and Penfold.[8] The low-temperature specific heat is 10.5 mJ/°C-mole corresponding to a density of states $\eta=2.23$ states/eV-atom. The susceptibility is strongly temperature-dependent rising to an extrapolated value of 3.20×10^{-4} emu/mole at $T=0$. Gardner and Penfold also quote a value of density of states calculated by Altman and Bradley which corresponds to $\eta_0=0.63$ states/eV-atom. In their analysis of these results, Gardner and Penfold assume an orbital susceptibility of 1.10×10^{-4} emu/mole yielding a spin susceptibility at $T=0$ of 2.10×10^{-4} emu/mole or 3.25 states/eV-atom. They then proceed to fit the spin susceptibility with Eq. (8) assuming $\eta_0=\eta$, or about 3.5 times the calculated value of Altman and Bradley. The fit requires $J=0.12$ eV and a narrow peak in the density of states at the Fermi level of width 0.10 eV. This procedure neglects any electron-phonon enhancement of the density of states.

If electron-phonon effects are present, η_0 can be much less than η. In that case stronger exchange effects are required to account for the high value of the susceptibility and less extreme variations of $\eta_0(E)$ with energy are required to account for the temperature dependence of $\chi(T)$. A good compromise between the experimental value $\eta=2.23$ states/eV-atom and the calculated value $\eta_0=0.65$ states/eV-atom would be to assume that the correct value of η_0 is about 1.3 states/eV-atom. This would mean that the density of states is enhanced by electron-phonon interaction by a factor $2.23/1.3=1.7$, and that a peak exists in the density of states about twice the value calculated by Altman and Bradley.

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[5] L. F. Mattheiss, Bull. Am. Phys. Soc. **9**, 251 (1964).
[6] K. Krebs, Phys. Letters **6**, 31 (1963).
[7] J. H. Wood, Phys. Rev. **126**, 517 (1962).
[8] W. E. Gardner and J. Penfold (to be published).