the Hall electric field in iron. From this empirical simulation, it would seem that the large peak in the thermoelectric power may arise from the spin-orbit interaction, as does the "spontaneous" Hall field.

At higher temperatures, agreement between the calculated $S(B, T)$ and S of iron is not expected to be too good, as critical fluctuations in the magnetization or shifts in the Fermi energies of the two bands[14] affect the thermoelectric power as the temperature approaches the Curie temperature.

At low temperatures, $S(B, T)$ is not affected very much by the field until the temperature is greater than approximately 100 K. Markov[11] and Goodings[12] have found that s-d transitions contribute to the thermoelectric power at low temperatures, but this contribution is small at the temperature of the peak in S of iron. Similarly phonon-drag and magnon-drag contributions are to be expected at low temperatures.

More rigorous investigations of the effect of the spin-orbit interaction on thermoelectromagnetic transport processes are underway.

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Magneto-Elastic Dependence of the Propagation of Sound in Gadolinium at the Critical Point*

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The change in the attenuation of sound and in the elastic constant c_{33} of gadolinium has been measured at 5 MHz as a function of applied magnetic fields to 13 kOe in the temperature range 270–320 °K. The minimum in the elastic constant at the Curie point moved upwards in temperature and tended to diminish and broaden with increasing field. Studies of the change in attenuation due to magnetic field in the same region revealed sharp λ-shaped increases up to a field of 1.8 kOe. At higher fields this peak split into two diminishing maxima, one moving upwards and the other downwards with increasing field. The zero-field-attenuation data decreased as $\Delta\alpha \sim \epsilon^y$, where $\epsilon = |(T - T_c)/T_c|$ and $y = -1.8 \pm 0.2$ in the paramagnetic region. In the presence of a magnetic field, the changes in the velocity are attributed to changes in the spin-correlation function.

INTRODUCTION

Anomalies in the sound attenuation and velocity at second-order phase transitions have been observed in many ferromagnetic materials. Near the Curie point, λ-shaped increases in the specific heat are normal and are accompanied by similarly shaped increases in the attenuation of sound as well as sharply peaked decreases in the sound velocity. These phenomena have been reported in nickel,[1-3] iron,[4] and in most of the rare earths including gadolinium.[5] The characteristic λ shape which the specific-heat and attenuation-vs-temperature curves assume is common to most ferromagnetic materials although there are some exceptions (e.g., EuO[6,7]). The specific heat of gadolinium exhibits the typical behavior[8] with its maximum near 290.2 °K.

The behavior of the velocity at the critical point is also typical. In the region around the Curie point decreases have been reported[5,9,10] with $\Delta v/v_0$ in the range of interaction assuming a maximum value of 12×10^{-3}. A sharp dip in c_{33} at 290.2 °K marks the Curie point and a broader minimum near 236 °K denotes the onset of the spin-rotation region.[9]

Lüthi *et al.*[5] have plotted $\Delta v/v_0$ vs ϵ for ferromagnetic rare earths at temperatures close to the Curie point. These results reveal a

$$\Delta v/v_0 \propto \tfrac{1}{2}\omega^0 \ln\epsilon \qquad (1)$$

dependence, where v_0 is a constant velocity above the range of interaction and ϵ is the reduced temperature $\epsilon \equiv |(T-T_c)/T_c|$ and ranges up to $(T-T_c) = 60$ °K. This type of result is obtained for critical-point behavior irrespective of whether the material is ferromagnetic or antiferromagnetic, isotropic or anisotropic.

Attenuation studies of gadolinium and other rare earth metals have also appeared in moderate numbers of late. Data have been reported by Rosen[11] for polycrystalline samples and by Lüthi and Pollina[12] and Lüthi *et al.*[5] for single crystals.

Above the Curie temperature the attenuation falls off as a power of the reduced temperature. Lüthi and Pollina[12] have reported a value of the critical exponent y of -1.2 ± 0.1 and Lüthi *et al.*[5] have reported a figure of -1.63 ± 0.1. The discrepancy is blamed upon an unusually anisotropic behavior in the first sample; thus these authors prefer the second value.

The frequency dependence generally goes as the square, leaving

$$\Delta\alpha(q,\epsilon) = Bq^2\epsilon^y , \qquad (2)$$

where q is the wave number. The temperature region over which this relation holds is reported as $1.5 < \Delta T < 15$ °K.

The simplicity of these relations is deceptive in that there are many delicately balanced relationships operating in this region. It was with the hope of examining these interrelationships that magnetic field experiments were undertaken.

EQUIPMENT AND SPECIMEN

A 99.9%-pure gadolinium specimen purchased from Alpha Inorganic Co. was oriented, annealed, and cut, as has been described by Long *et al.*[9] Its final configuration was a right cylinder 5 mm in diameter and 3.091 mm long with its axis parallel to the unique axis of the hexagonal crystal. Its end faces were cut on a Materials Research Ltd. Servomet spark cutter and the orientation of the unique axis was determined on a General Electric XRD-5 x-ray spectrometer to within ±1°.

Throughout the entire course of the experiment one set of transducers and one bond were employed. $\frac{1}{8}$-in.-diam x-cut quartz transducers with a 5-MHz resonant frequency and an overtone polish were

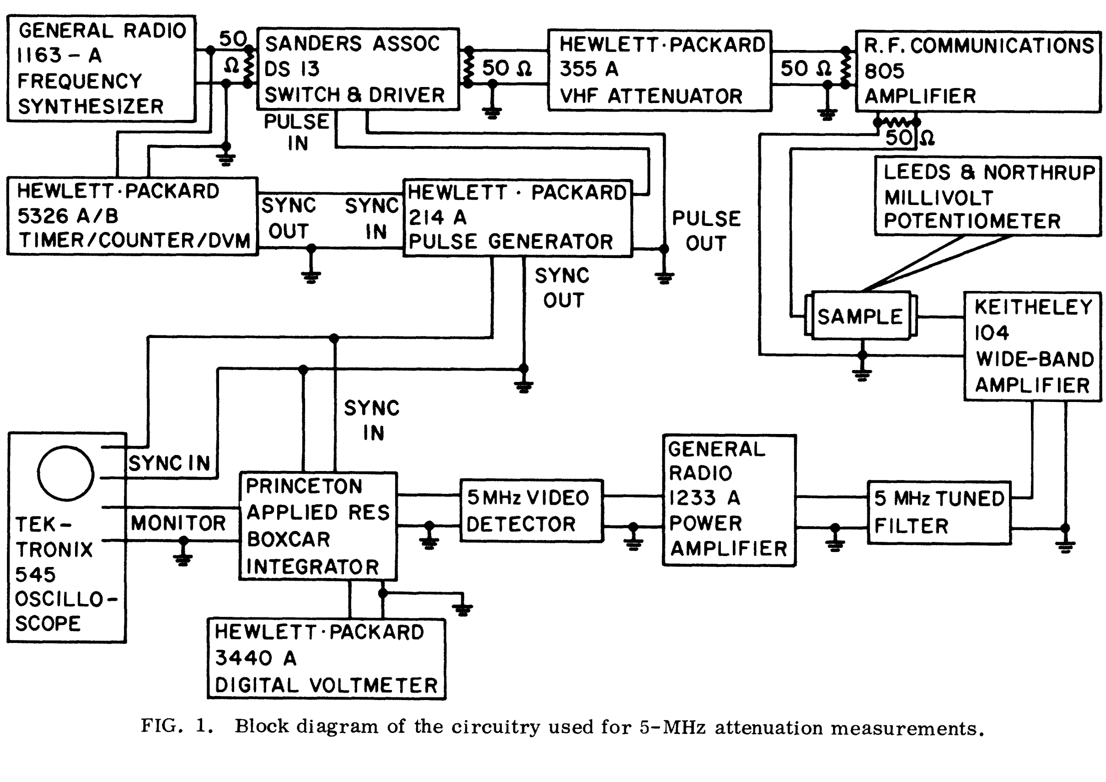

FIG. 1. Block diagram of the circuitry used for 5-MHz attenuation measurements.

bonded to a sample's faces with Armstrong C-2 Epoxy and an E activator as described by Levy and Rudnick.[13]

The dc magnetic field was produced by a Varian model V 4012 12-in. electromagnet and a model V 2100-A regulated power supply and controlled by a Mk. I fieldial regulator. Pole pieces were tapered to a minimum diameter of $7\frac{1}{2}$ in. at a gap spacing of $2\frac{1}{2}$ in. The highest field available was about 13 kOe.

Temperature control was maintained by a cold-gas system[14] which could hold the sample temperature to at least 0.1 °K for periods of up to several hours. Liquid nitrogen was boiled and the gas blown through a heat exchanger and over the sample. The output from a sensing thermocouple was fed back to regulate the power to the heat exchanger

Velocity measurements were made following the method of Williams and Lamb[15] with a circuit similar to that discussed by Long *et al.*[9] Attenuation measurements were made with the circuit configuration shown in Fig. 1.

EXPERIMENTS AND RESULTS

Measurements of the elastic constant c_{33} and the ultrasonic attenuation at 5 MHz have been made in the region around the Curie temperature, with and without applied magnetic fields of up to 13 kOe.

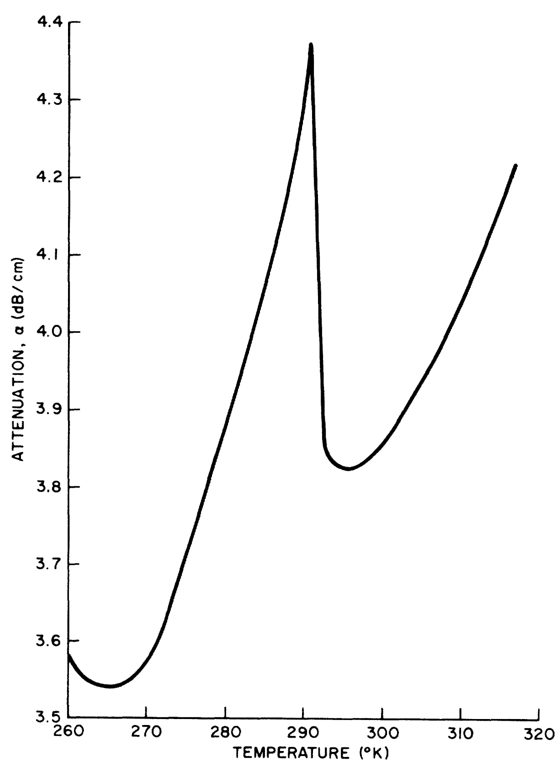

FIG. 2. Absolute attenuation α vs temperature for 5-MHz c_{33} waves in gadolinium.

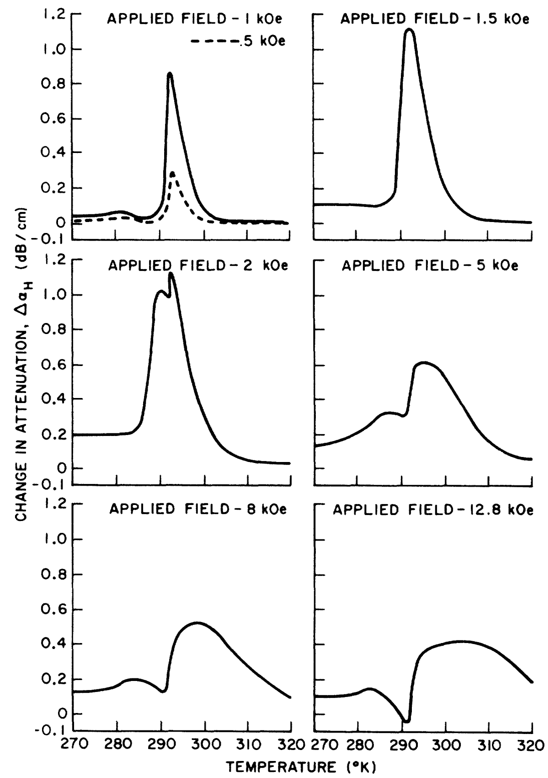

FIG. 3. Change in attenuation with unique-axis applied magnetic fields, $\Delta\alpha_H$, vs temperature for 5-MHz c_{33} waves in gadolinium.

The elastic constants were calculated from measurements of the velocity according to

$$c_{33} = \rho v^2_{[001]} \tag{3}$$

using $\rho = 7.895$ g/cm^3.

Absolute attenuation measurements were made by measuring the peak height of the first four ultrasonic echos with a boxcar integrator. The attenuation was then calculated from

$$\alpha = \Delta\alpha + \alpha_B = \frac{1}{3}\sum_{n=2}^{4}\frac{20}{n(2l_s)}\log_{10}\frac{\langle P_n\rangle}{\langle P_1\rangle}, \tag{4}$$

where l_s is the sample length and $\langle P_n\rangle$ the average height of the nth pulse. The background attenuation α_B was estimated by fitting a baseline curve to the data and then subtracting it off to give $\Delta\alpha$.

The usual λ shape was observed at the Curie point. The absolute attenuation data shown in Fig. 2 showed agreement to within the experimental error with the results of Lüthi *et al.*,[5] fitting an inverse power law in the temperature range $7 > \Delta T > 1$ °K:

$$\Delta\alpha/\omega^2 \sim \epsilon^y, \tag{5}$$

with $y = -1.8 \pm 0.2$. Errors in the evaluation of y were mostly due to the difficulty in estimating back-

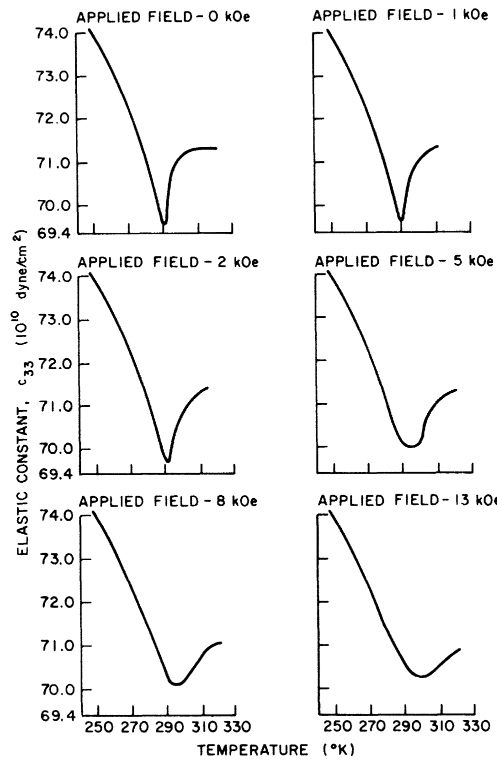

FIG. 4. Elastic constant c_{33} vs temperature for unique-axis applied magnetic fields at 5 MHz in gadolinium.

ground levels, which were not linear at these frequencies.

Figure 3 shows the additional attenuation $\Delta\alpha_H$ due to an applied magnetic field along the unique axis of the crystal. The maximum attenuation was recorded for a field of about 1.8 kOe at the Curie point. Two maxima were observed above this field as well as a general broadening of the temperature range of the interaction. The temperatures at which the two maxima occurred became increasingly separated with increasing field. The change in attenuation due to magnetic field was obtained by repeated measurements of the first echo pulse with and without magnetic field. A calculation similar to that of Eq. (3) gave the change in attenuation, $\Delta\alpha_H$.

The movement of the attenuation peaks prompted the elastic constant measurements in this range. These resulted in the curves shown as Fig. 4. Here also there is an apparent shift in the Curie temperature upward with increasing field as well as a general broadening of the elastic constant minimum with temperature. Plotting the temperature at which the minimum in the elastic constant occurs vs magnetic field in Fig. 5, we find a

$$T_c'(H) = T_c + CH \tag{6}$$

behavior at high fields with $C = 0.77$. At low fields the data are not linear.

Finally, Fig. 6 shows the change in attenuation, $\Delta\alpha_H$, vs magnetic field at the zero-field Curie temperature. No account has been taken of the demagnetization factor in the presentation of these data.

DISCUSSION

Recent theoretical work has isolated the dominant terms in the total ferromagnetic Hamiltonian given by Long *et al.*[9] Freyne,[16] using a molecular-field approach, has truncated it to

$$\mathcal{H} = -g\mu_B \vec{H}_e \sum_i \vec{S}_i + \tfrac{1}{2}c_{11}^{\alpha}(\varepsilon_1^{\alpha})^2 + c_{12}^{\alpha}\varepsilon_1^{\alpha}\varepsilon_2^{\alpha} + \tfrac{1}{2}c_{22}^{\alpha}(\varepsilon_2^{\alpha})^2 - \tfrac{1}{2}\sqrt{3}\,[\tilde{\beta}_{12}^{\alpha}\varepsilon_1^{\alpha} + \tilde{\beta}_{22}^{\alpha}\varepsilon_2^{\alpha}]\,[S_z^2 - \tfrac{1}{3}S(S+1)]$$
$$- \tfrac{3}{2}P_2[S_z^2 - \tfrac{1}{3}S(S+1)] - \tfrac{1}{8}P_4[35S_z^4 - 30S_z^2 + 3S(S+1)] - \tfrac{1}{16}P_6[231S_z^6 - 315S_z^4 + 105S_z^2 - 5S(S+1)]\,. \tag{7}$$

In calculating the velocity of sound from this Hamiltonian they have demonstrated that the inclusion of the terms

$$\mathcal{H}_M = -\tfrac{1}{2}\sqrt{3}\,(\tilde{B}_{12}^{\alpha}\varepsilon^{\alpha,1} + \tilde{B}_{22}^{\alpha}\varepsilon^{\alpha,2})\,[(S_i^z)^2 - \tfrac{1}{3}S(S+1)]\,, \tag{8}$$

where

$$\varepsilon^{\alpha,1} = \varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33}\,,\qquad \varepsilon^{\alpha,2} = \tfrac{1}{2}\sqrt{3}(\varepsilon_{33} - \tfrac{1}{3}\varepsilon^{\alpha,1}), \tag{9}$$

is sufficient to characterize the velocity changes accompanying the spontaneous realignment of spins which occurs in the spin-rotation region near 236 °K. Since the $\tilde{B}_{ij}^{\alpha}$ coefficients are slowly varying with temperature it is reasonable to assume that these same terms will dominate the magnetoelastic Hamiltonian near the Curie point. Callen and Callen[17] have included the temperature and field dependence of the magnetostrictive terms in the correlation function

$$\mathcal{L}_f(T, M) = \langle (S_f^z)^2 - \tfrac{1}{3}S(S+1)\rangle\,. \tag{10}$$

In the vicinity of the Curie point the application of a magnetic field should retard the drop in the spin-correlation function and shift the Curie temperature upwards. By this mechanism the application of a magnetic field should diminish the magnitude of the drop in the velocity as well as broadening the range of interaction. This is, in fact, what is observed.

There has recently been some discussion concerning the behavior of the heat capacity of gadolinium near the critical point. Lewis[18] has found a rounded peak in the λ-shaped heat capacity, which led him to question its "ideal" behavior. It is generally accepted that the anomaly in the velocity,

$$-\Delta v/v_0 \sim \omega^0 C_v \ , \tag{11}$$

has the same character as that in the specific heat.[19, 20] The velocity data of this work are less rounded than that of Lewis ($T_c \pm \frac{1}{2}$ °K), but it is unclear whether this rounding is due to thermal gradients, residual field, or departure from "ideal" behavior. The zero-field measurements of Long *et al.*[9] were made in a residual field of the order of the earth's field and showed no rounding to within ±0.2 °K.

With the application of a magnetic field the velocity curve is definitely rounded and broadened. One might expect the same behavior of the specific heat.

The attenuation effects are somewhat more complex. Laramore and Kadanoff[21] have derived an expression for the behavior of the attenuation of sound of a ferromagnet in the paramagnetic region in the absence of a magnetic field:

$$\Delta\alpha \sim \frac{q^2 v^3 \rho k_B T_c \gamma^2}{\xi_0^3}\left(\frac{1}{T_c}\frac{dT_c}{d\sigma}\right)\frac{\epsilon^y}{m_s^*} \ , \tag{12}$$

where ρ is the density, k_B the Boltzmann constant, ξ_0 the critical-point correlation length, σ the stress, m_s^* the resonant frequency of a typical spin fluctuation, and y and γ critical exponents. This calculation predicts the attenuation

$$\Delta\alpha \sim q^2 \epsilon^{-5/3 + 11\alpha/6 + \eta/3 - \alpha\eta/6} \tag{13}$$

for gadolinium. Lewis[18] gives $\alpha = -0.09 \pm 0.05$ and

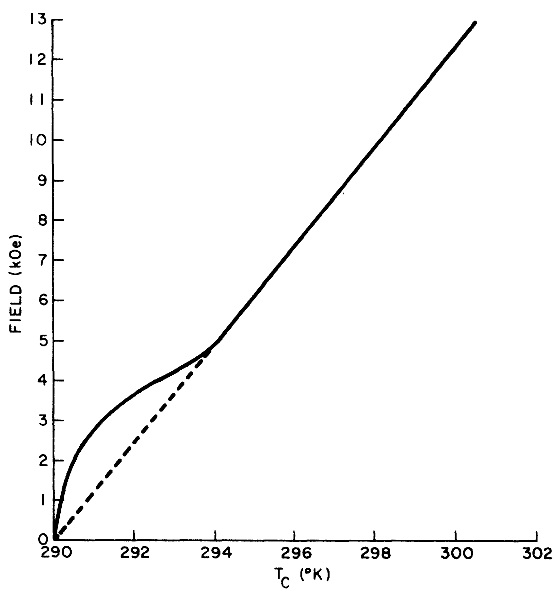

FIG. 5. Apparent Curie temperature vs applied unique-axis magnetic field in gadolinium.

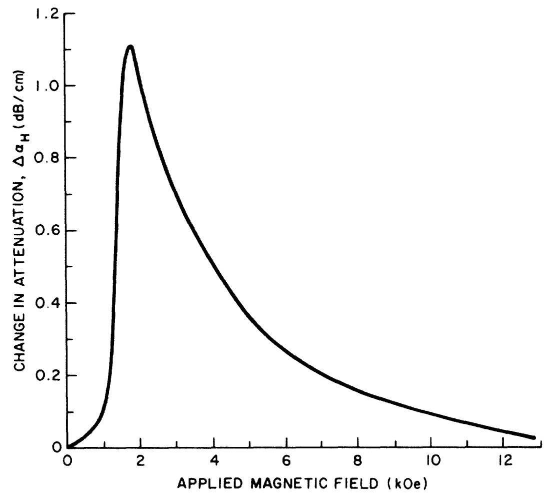

FIG. 6. Change in attenuation of 5-MHz c_{33} waves, $\Delta\alpha_H$, vs applied unique-axis magnetic field at 290.2 °K.

Kadanoff *et al.*[22] give $\eta = 0.07 \pm 0.07$, which yields

$$\Delta\alpha \sim q^2 \epsilon^{-1.81} \ , \tag{14}$$

in excellent agreement with our measured value of $y = -1.8 \pm 0.2$.

Since the expression of Laramore and Kadanoff gives the closest theoretical prediction, it may serve as a jumping-off point for the analysis of the magnetic field behavior. The mechanism of absorption in the paramagnetic region postulated by Laramore and Kadanoff is the variation in the magnetic-order parameter due to the creation of two spin fluctuations at the expense of a phonon. In the presence of a magnetic field, the increase in the spin-correlation length will increase $\Delta\alpha$ through m_s^* and broaden the range of interaction. This is consistent with the behavior of the velocity. It is not clear that this mechanism alone can account for the rather large changes in attenuation in the paramagnetic region.

The origin of the phenomenon of the double attenuation peak is also unclear. Robinson *et al.*[23] have shown that one obtains a decrease in the Curie temperature with increasing pressure in gadolinium. It is conjectured that the cause of this effect may be contributory to the motion of the lower peak.

Further experimentation is clearly desirable. It is hoped that those capable of extending the range of the magnetic fields applied in these experiments will do so. Where possible, simultaneous measurements of the Curie point should be made in order to separate multiple effects.

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Kondo-Resistivity Suppression Due to Inelastic Scattering of Conduction Electrons by Magnetic Impurities in Alloys

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The interaction of localized magnetic moments of a magnetic impurity with the lattice causes relaxation of the states of that localized moment. The lifetime is represented as a broadening γ of the states of the localized moment. The introduction of this broadening changes the scattering of the conduction electrons by the localized moment from an elastic scattering to an inelastic one plus negligible elastic scattering. Thus, the electron of the average excitation energy above the Fermi level $\epsilon \approx T$ may take part in all inelastic allowed scattering processes only if $\epsilon \gtrsim \gamma$, i.e., $T \gtrsim \gamma$. For $T < \gamma$ this number of processes is reduced, and it vanishes for $T \rightarrow 0$. This effect was in fact found for the terms of the order J^2 and J^3 in the perturbation expansion, and the correct temperature dependence of the Kondo resistivity was obtained for $J < 0$. The resistivity for $J > 0$ is also discussed.

Recent measurements of the Kondo resistivity of Cu-Mn,[1] Au-Mn,[2] Ag-Mn,[3] and Au-Fe[4] alloys have shown deviation from the Kondo $1 - A\ln(\epsilon_F/T)$ behavior for low temperatures. It was found that at low temperatures the resistivity due to magnetic impurities first increases until it reaches a maximum (at T_M) and then decreases again and can be described by a $1 - A\ln(\epsilon_F/T)$ law for $T \gg T_M$. The Kondo temperature (T_K) for the Cu-Mn, Au-Mn, Ag-Mn systems is very low ($T \ll 0.1\,^\circ$K) and for Au-Fe it is[4] $T_K = 0.24\,^\circ$K. In all these cases[1-4] T_K is much smaller than T_M. Since $T_K \ll T_M$, the maximum in the resistivity cannot be associated with the formation of a quasibound state of conduction electrons around an impurity spin for $T < T_K$.[5] Thus a different mechanism, which suppresses the Kondo resistivity at $T < T_M$, must be looked for. Such a mechanism was suggested by Harrison *et al.*[6] They assume that the existence of an internal local magnetic field due to Ruderman-Kittel-Kasuya-Yosida (RKKY) impurity-impurity interaction suppresses the Kondo resistivity.[7] Silverstein[8] has considered the change of the mean magnetic moment of the impurity spin due to the change of the occupation of the Zeeman levels with the change in temperature in this internal local magnetic field. The calculated resistivity is in quite good agreement with experiment for T around T_M.[2-6] We, however, prefer to attribute to the RKKY impurity-impurity interaction merely a