communicating their results prior to publication.

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IMPURITY NUCLEAR-MAGNETIC-RESONANCE SHIFTS AND SPIN-LATTICE RELAXATION RATES IN Al:V, Al:Cr, and Al:Mn†

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Low-temperature (1.3-4.0°K) nuclear resonance shifts and spin-lattice relaxation rates of dilute "nonmagnetic" ^{51}V, ^{53}Cr, and ^{55}Mn impurities in aluminum are compared against similar data for the "magnetic" alloys Au:^{51}V and Mo:^{59}Co. It is shown that no essential differences exist between the two types of alloy systems.

A sharp phase boundary between magnetic and nonmagnetic regimes occurs in the Hartree-Fock treatment of Anderson's model[1] of dilute transition-element impurities in metals. Recent evidence[2,3] suggests, however, that the distinction between the two regimes is largely removed if dynamic effects are included in the treatment of the problem. In this approach a localized moment is viewed quite generally as a localized spin fluctuation[4] whose lifetime is proportional to $[1-UN_d(0)]^{-1}$, where U is the Coulomb repulsion between electrons of opposite spin on the impurity and $N_d(0)$ is the density of virtual d states at the Fermi level. Thus, a gradual transition from nonmagnetic to magnetic behavior occurs as the fluctuation lifetime increases. In the magnetic limit, which corresponds to an infinite lifetime [i.e., $UN_d(0) \geq 1$], the properties of the localized moment are usually discussed in terms of an effective s-d exchange interaction $-J\vec{S}\cdot\vec{\sigma}$ between the impurity d spin $\vec{S}$ and the conduction-electron spins $\vec{\sigma}$. At sufficient low temperatures and $J<0$ this interaction leads to anomalous transport properties and spin compensation of the impurity moment as a result of the well-known divergence[5] of the conduction-electron impurity-scattering amplitude below the Kondo temperature (T_K). It has been claimed[3,4] that conduction-electron scattering from localized spin fluctuations gives rise to similar anomalies. This would imply that the distinction between magnetic and nonmagnetic regimes is artificial, thus making it impossible to infer the sign of $[1-UN_d(0)]$ from the presence or absence of specific low-temperature anomalies such as the resistance minimum. However, with the possible exception of the behavior of iron impurities in copper-zinc alloys[6] no direct experimental support exists for this conjecture.

The purpose of this note is to report low-temperature measurements[7] of impurity nuclear-magnetic-resonance (NMR) shifts (K) and spin-lattice relaxation rates (T_1^{-1}) in the "nonmagnetic" dilute alloys Al:V, Al:Cr, and Al:Mn,[8] and to compare the results with similar measurements for the "magnetic" alloys Au:V[9] ($T_K \approx 300°\mathrm{K}$)[10,11] and Mo:Co[12] ($T_K \approx 24°\mathrm{K}$).[13,14] The NMR shifts and relaxation rates are determined by the local static and dynamic susceptibilities, respectively, and thus provide a microscopic probe of the magnetic response of the impurity. The notation "magnetic" and "nonmagnetic" refers to the models (i.e., Kondo or spin fluctuation) which have been most successful to date in accounting for the bulk properties of the respective alloys. It is difficult, of course, to obtain independent, reliable estimates of the important parameters, such as $UN_d(0)$, which enter into the magnetic-impurity problem. For this reason one generally cannot determine uniquely whether a given alloy is magnetic or nonmagnetic in the traditional Friedel-Anderson sense. This uncertainty, however, is of little consequence here since our aim is to explore possible qualitative differences in the local magnetic properties of a representative group of alloys whose bulk properties reflect the wide range of parameter values normally encountered.

All measurements were carried out on 200-300 mesh powders in the temperature range 1.3-4.0°K by means of standard spin-echo techniques. Adequate signal-to-noise ratios were achieved either with a Princeton Applied Research Model No. CW-1 boxcar integrator or a Fabri-Tek 952/1062 high-speed digital signal averager. Magnetic fields to 60 kOe were produced in a superconducting NbZr solenoid. The alloys were prepared in an inert-gas arc furnace. The impurity concentrations were 0.1 and 0.5 at.% for the vanadium and chromium (enriched to 96% ^{53}Cr) alloys, respectively. In the case of Al:Mn several concentrations in the range 0.04-0.3 at.% were studied. No concentration dependence was observed in either the resonance shifts or relaxation rates.

Table I. Summary of impurity NMR data for Al:V, Al:Cr, and Al:Mn.

	K (%)	T_1T (sec °K)
^{51}V	+(0.30 ±0.03)	0.44 (±0.03)
^{53}Cr	−(0.38 ±0.03)	1.55 (±0.30)
^{55}Mn	−(2.01 ±0.05)	0.019 (±0.003)

Our experimental results are summarized in Table I.[15] The negative resonance shifts for ^{53}Cr and ^{55}Mn can be attributed to a d-spin core-polarization hyperfine interaction. The dominance of this mechanism is a consequence of the high virtual d-state density[8] in these alloys. Moreover, the variation of the observed shifts is qualitatively consistent with the increasing bulk susceptibility[8] in the sequence Al:V, Al:Cr, and Al:Mn. In the temperature range of our experiments the spin-lattice relaxation rates are directly proportional to the absolute temperature, as illustrated in Fig. 1 for Al:Mn. The observed temperature dependence of T_1^{-1} is characteristic of hyperfine coupling mechanisms involving itinerant electrons and is related to the thermal broadening of the Fermi level (i.e., the rate is proportional to the average number of unpaired conduction-electron spins).

The relatively large negative resonance shifts and rapid relaxation rates of ^{53}Cr and ^{55}Mn differ markedly from the corresponding quantities in typical paramagnetic transition metals such as vanadium[16] ($K = +0.58\,\%$ and $T_1T = 0.79$ sec °K).

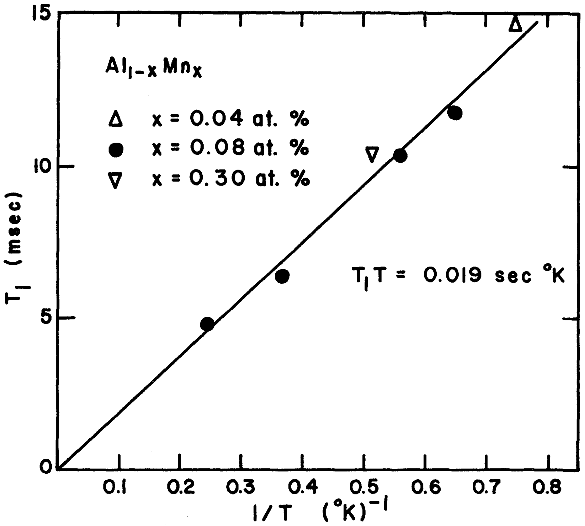

FIG. 1. Plot of ^{55}Mn spin-lattice relaxation times as a function of reciprocal temperature for three different Al:Mn alloy compositions.

Table II. Comparison of impurity NMR results. The impurity susceptibilities $\Delta\chi$ are extrapolated 0°K values.

	$\Delta\chi$ (10^{-4} emu/g atom)	$\gamma_n^2T_1T$ (10^6 sec^{-1} °K Oe^{-2})	K (%)	K_d (%)	K_{VV} (%)
Al:^{51}V	-4^a	28	+0.30	−0.7	+1.0
Al:^{53}Cr	7^a	3.5	−0.38	−1.6	+1.2
Al:^{55}Mn	14^a	0.82	−2.0	−3.5	+1.5
Au:^{51}V	47^b	0.84^c	-1.5^c	−3.3	+1.8
Mo:^{59}Co	290^d	0.10^e	-7.3^e	−9.5	+2.2

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This difference reflects the smaller d-level width in the aluminum alloys. The resonance shift is proportional to the static susceptibility, which is enhanced by the factor $[1-UN_d(0)]^{-1}$. Because of the local character of the enhancement mechanism the low-frequency transverse susceptibilities and hence the spin-lattice relaxation rate[17] are enhanced by the factor $[1-UN_d(0)]^{-2}$. It follows that the core-polarization shift and relaxation rate are related by the Korringa-like relation

$$K_d^2T_1T = 5(\gamma_e/\gamma_n)^2(\hbar/4\pi k_B), \tag{1}$$

where the various quantities have their usual meaning and K_d is taken to be negative. The factor 5 in Eq. (1) arises from the fivefold orbital degeneracy of the d states.[18] We have made the assumption that the fractional weights of the five d orbitals at the Fermi level are equal. Applying the modified Korringa relation to our relaxation data yields estimates for the core-polarization shifts which are listed in Table II. In each of the three aluminum alloys the calculated shift is more negative than the observed shift. The difference can be attributed to a positive orbital contribution (K_{VV}) whose magnitude is proportional to the product of a coupling constant ($\langle r^{-3}\rangle$) and the orbital susceptibility of the impurity. For a given occupation number, the latter should scale with the reciprocal d-level width. The values of K_{VV} ($=K-K_d$) listed in Table II are consistent with the known orbital shift of ~+0.6% in vanadium metal.[16] Since the orbital relaxation rate is not related to K_{VV} by a Korringa-like relation, its magnitude cannot be estimated reliably. However, in view of the reasonable magnitudes of K_{VV} inferred from the NMR data it appears that the orbital relaxation mechanism is relatively unimportant. The direct 4s contact interaction can also be ignored since its effect is unlikely to be significantly greater than in vanadium metal ($K_s \approx +0.1\%$). Moreover, any 4s contribution resulting from local s-d exchange polarization effects can be included in the d-spin core-polarization hyperfine coupling constant and thus does not affect our analysis.

We now turn to a comparison of the aluminum alloy results with those for Au:V and Mo:Co. A summary of the available NMR data together with the 0°K impurity susceptibilities ($\Delta\chi$) is given in Table II. The ^{51}V and ^{59}Co data are noteworthy in two respects. In the first place, the spin-lattice relaxation rates are again proportional to the absolute temperature, suggesting that the relaxation mechanism is basically the same as in the "nonmagnetic" alloys. Secondly, the relaxation rates and resonance shifts can be related as in the "nonmagnetic" alloys by the modified Korringa relation. The resulting orbital shifts have magnitudes which are slightly larger than those inferred for the aluminum alloys in agreement with the expected decrease in the virtual level widths.

The only distinguishing feature between the "magnetic" and "nonmagnetic" alloys considered here is the magnitude of the d-spin core-polarization hyperfine field. Assuming that the measured impurity susceptibility is localized in every case on the impurity site and ignoring orbital contributions to $\Delta\chi$ we obtain hyperfine fields according to $H_{\text{hfs}}^{(d)} = \mu_BNK_d/\Delta\chi$ (where μ_B is the Bohr magneton and N is Avogadro's number) which have nearly the free-ion value in Al:Cr (-130 kOe/μ_B) and Al:Mn (-140 kOe/μ_B), but are much smaller in Au:V (-39 kOe/μ_B) and Mo:Co (-18 kOe/μ_B). Although the significance of these results is not clear it is likely that they simply reflect differences in host properties.

In conclusion, the temperature independence of T_1T and the consistent behavior of K^2T_1T for dilute alloys spanning a wide range of 0°K susceptibilities indicates that the magnetic properties of alloys previously treated separately as spin-fluctuation and Kondo systems are qualitatively indistinguishable at sufficiently low temperatures. Moreover, the NMR results appear to be easily understood in terms of a single-particle description. This observation is consistent with the linear temperature dependence of the low-temperature specific heats of Au:V[19] and Cu:Fe.[20]

It is a pleasure to acknowledge several helpful conversations with L. Dworin.

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INFLUENCE OF SMALL-ANGLE SCATTERING ON OPEN-ORBIT CONDUCTION IN THALLIUM*

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A measurement of the electrical and thermal magnetoresistance of thallium gives a direct demonstration of the scattering of open-orbit electrons into adjacent closed orbits through the mechanism of small-angle scattering. Potentially, this effect provides a method for determining the efficiency of small-angle scattering in metallic conduction.

In a metal, both electrical and thermal resistance depend greatly on how close an electron is brought to the equilibrium distribution after being scattered. Even though small-angle scattering would be efficient enough to cause thermal resistance, it is not generally expected to cause appreciable resistance to an electrical current. There are known exceptions, viz., the influence of small-angle scattering on the galvanomagnetic effects in metals has been discussed recently by several authors.[1] Of these, Pippard's description of the expected behavior of the transverse magnetoresistance due to open-orbit electrons being scattered into adjacent closed orbits by small-angle scattering is the most appropriate for the work reported here.

In thallium a slice of open orbits exists on the honeycomblike fourth-zone electron sheet of the Fermi surface[2] when a magnetic field is applied close to the hexagonal direction. The narrow re-