No resonance was observed in a silver-1 at.% dysprosium alloy, but at 4.2°K a strong resonance was seen in a silver-0.8 at.% gadolinium alloy, with $g = 2.005 \pm 0.005$ and $\Delta H = 158$ G; this was still visible at 85°K with $\Delta H = 510$ G. This g value is in good agreement with the results of earlier work[6]; it is not yet clear whether the considerable smaller linewidth is due to the absence of low-temperature magnetic ordering[11] in our appreciably more dilute alloy or to differences in the metallurgical character of the specimen.

Dilute alloys of transition metals in copper or silver carry localized moments which are associated[12] with virtual bound states, and related behavior is found[13,14] for iron in some 4*d* transition elements. Rare-earth solutes in simple metals must possess considerably narrower virtual bound states, and as the solute approaches the end of the 4*f* series a truly localized state is likely to form below the bottom of the conduction band of the solvent. We suggest that our observation of a non-*S*-state resonance for erbium has been made possible by the formation of such a state, and that for earlier rare-earth elements in metallic solvents, the broadening of the virtual states mixes components of different g value too much for any resonance except that of Gd^{+++} ($^8S_{7/2}, g = 2$) to be observed.

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GIGANTIC MAGNETIC SUSCEPTIBILITIES IN PARAMAGNETIC SYSTEMS*

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Often, metals with small amounts of dissolved transition-metal or rare-earth impurities (~2-5 at.%) exhibit a peak in the paramagnetic susceptibility at low temperatures.[1] In the past this has usually been interpreted as an indication of the onset of antiferromagnetism, since this behavior is characteristic of the idealized two-sublattice Ising-model antiferromagnet.[2] Recently, Klein,[3,4] in an extension of the Marshall-Brout-Klein[5] theory of metallic systems with random dilute magnetic impurities, demonstrated the possibility of the existence of a peak in the magnetic susceptibility in a metal with disordered spins, in the absence of long-range order (ferromagnetism or antiferromagnetism). We have examined the magnetic behavior of $La_{3-x}Gd_xIn$, an intermetallic compound with the $L1_2$, Cu_3Au, cubic crystal structure.[6] Its behavior is characterized by some features which are common to other metallic paramagnetic systems which have been studied. For example, there is a peak in the magnetic susceptibility as a function of temperature, and the temperature at which the peak occurs,

T_{max}, is proportional to the concentration of spin impurities (for low concentrations of Gd). However, the behavior of the $La_{3-x}Gd_xIn$ system is also characterized by new and surprising features which cannot be explained in terms of the Brout-Klein theory[3-5] or other current theories. These are, first, that the paramagnetic susceptibility at the susceptibility maximum is exceedingly large compared to the calculated free-ion susceptibility, even though the system, for low Gd concentration, exhibits no magnetic remanence and therefore no long-range ferromagnetic order at temperatures well below T_{max}; and second, that the magnitude of the susceptibility at the peak depends strongly on the concentration of Gd spins.

The $La_{3-x}Gd_xIn$ samples were prepared by melting together the constituents in a conventional arc furnace. The resulting ingots were then machined into cylindrical samples approximately 3 mm in diameter and 4 cm long. The magnetization was measured by withdrawing the sample from a pickup coil (which was connected in series to a ballistic galvanometer) in the presence of a magnetic field whose direction was along the axis of the sample. The sensitivity of the apparatus was such that the magnetization of the more dilute samples could be measured in an applied magnetic field of a few oersteds. The zero-field paramagnetic susceptibilities were determined from the slope of the curves of magnetization versus magnetic field, in the limit of zero magnetic field. The apparatus was calibrated by measuring the known susceptibility of a manganous ammonium sulfate sample.

The zero-field magnetic susceptibilities, normalized to the free-ion Brillouin susceptibilities, for $La_{3-x}Gd_xIn$ alloys with Gd concentrations of 2.61, 3.00, 4.26, 5.76, and 6.53 at.% and for a La-Gd alloy with 5.0 at.% Gd are shown in Fig. 1. When the magnetic field was reduced from 1000 G to zero at 1.2°K, the 2.61 and 3.00% Gd samples had no magnetic remanence, the 4.26% sample had 2% remanence (2% of the 1000-G field) and the 5.76 and 6.53% samples had approximately 10% remanence. The La-5% Gd sample had 20% remanence at 1.2°K. From this we can say that the $La_{3-x}Gd_xIn$ system, for Gd concentrations up to approximately 4 at.%, exhibits no long-range ferromagnetic order at temperatures well below T_{max} (insofar as the absence of remanence implies the lack of long-range magnetic order). For the 5.76 and 6.53% Gd systems the small remanence values suggest there is at least some long-range ferromagnetic order. As shown in Fig. 2, T_{max} is proportional to the Gd concentration at lower concentrations but increases somewhat more rapidly than linearly with increasing concentration at concentrations higher than approximately 4 at.%. The magnetic susceptibility (normalized to the free-ion value) at the susceptibility maximum closely follows the relationship $\chi_{max}/\chi_{B\ max} \propto n^{7/2}$, where $\chi_{B\ max}$ is the free-ion Brillouin susceptibility at T_{max} and n is the Gd concentration.

Klein[4] extended the Marshall-Brout-Klein theory to calculate the behavior of the electric

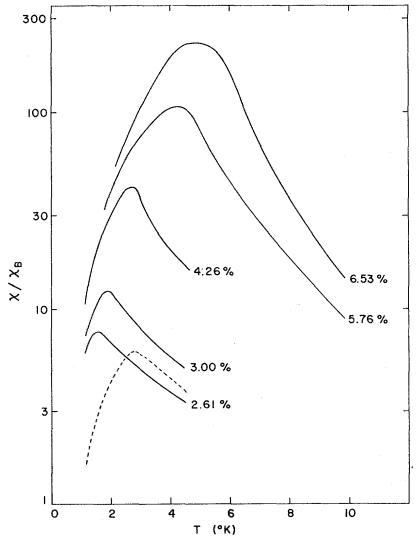

FIG. 1. Magnetic susceptibilities (normalized to the free-ion value χ_B) versus temperature for five $La_{3-x}Gd_xIn$ samples (solid lines; atomic percent Gd is indicated on graph) and a La-5% Gd sample (dashed line), plotted on a semilog grid.

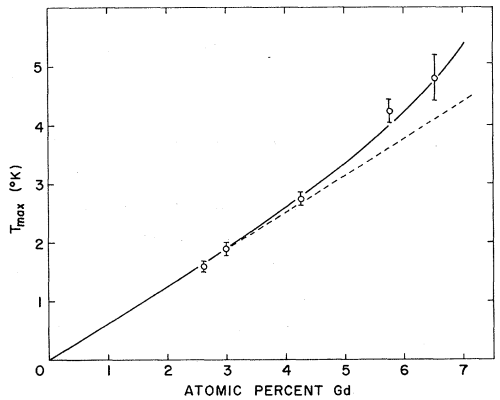

FIG. 2. The temperature of the susceptibility maximum (from Fig. 1) as a function of atomic percent Gd for the $La_{3-x}Gd_xIn$ system.

susceptibility of a random dilute ferroelectric system. Because of the generalized nature of the treatment, it is expected that the same theory should describe the paramagnetic susceptibility of a random dilute paramagnetic system[7] such as $La_{3-x}Gd_xIn$ (for sufficiently small values of x). The underlying basis of the theory is that the spin correlation between widely separated spin impurities (or the correlation between widely separated electric dipoles in the ferroelectric case) is broken up by randomly positioned impurities between them. There are small strongly correlated clusters of spins which are only weakly coupled to other clusters. Klein[4] has considered in detail only the case where the forces between the impurities are limited to dipole-dipole interactions, for which he obtains a temperature maximum in the susceptibility and the prediction that T_{max} is proportional to the concentration of impurities, but that the height of the peak is independent of the concentration and the susceptibility is smaller than the free-ion susceptibility at all temperatures.

It is conceivable that the strong concentration dependence of T_{max} and the gigantic magnetic susceptibilities of the $La_{3-x}Gd_xIn$ system can be brought into agreement with the Marshall-Brout-Klein theory if strong short-range ferromagnetic forces are included in the theory. On the other hand, it may be that the theory is inapplicable to systems with impurity concentrations as high as those reported here.

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MULTIPLE SPIN ECHOES AND SPIN LOCKING IN SOLIDS

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It is commonly believed that spin-echo techniques[1] succeed in recovering transverse spin magnetization $\gamma\hbar\langle I_x\rangle$ only to the extent that it has been lost through inhomogeneous broadening. We were, therefore, surprised to observe long trains of echoes in a number of homogeneously broadened solids when pulse sequences timed according to the Carr-Purcell prescription[2] were applied. (An initial 90° pulse is followed after a time τ by a train of pulses of repetition period 2τ.) An example is shown in Fig. 1. The experiments were all performed near room temperature with a pulse spectrometer which provided a rotating field $H_1 \approx 50$ Oe at 30 MHz and recovered from pulse overloads in ~5 μsec.

The behavior of this effect can be described as follows: (1) The phase of the rf carrier in the first pulse must differ by ~90° from that during the rest of the experiment.[3] (2) The effect occurs only when $\tau \lesssim T_2$. The echo en-

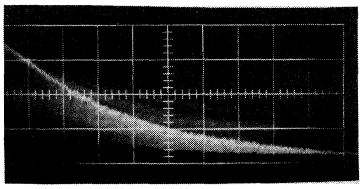

FIG. 1. Train of ^{19}F echoes in powdered $Co(NH_3)_6(BF_4)_3$. Sweep speed: 1 msec/cm. $\tau = 10$ μsec. For this substance $T_2 \approx 45$ μsec.