of the gallium droplets provided this is below about 100 μm. L. Bosio, thesis, Faculty of Sciences at the University of Paris, 1965 (unpublished).

[6] L. Bosio, A. Defrain, H. Curien, and A. Rimsky, Acta Crystallogr., Sect. B 25, 995 (1969).

[7] A. Bererhi, L. Bosio, R. Cortes, and A. Defrain, Electrodeposition Surf. Treat. 2, 455 (1974), and J. Phys. (Paris), Colloq. 36, C2-79 (1975).

[8] L. Bosio, R. Cortes, and A. Defrain, J. Chim. Phys. 70, 357 (1973).

[9] W. Marshall and S. W. Lovesey, *Theory of Thermal Neutron Scattering* (Clarendon Press, Oxford, 1971); J. R. D. Copley and S. W. Lovesey, Rep. Prog. Phys. 38, 461 (1975).

[10] D. I. Page, D. H. Saunderson, and C. G. Windsor, J. Phys. C: Solid State Phys. 6, 212 (1973).

[11] M. I. Barker, M. W. Johnson, N. H. March, and D. I. Page, in *Properties of Liquid Metals: Proceedings,* edited by S. Takeuchi and S. Takeuchi (Halsted Press, New York, 1973), p. 99.

[12] S. H. Chen, Y. Lefevre, and S. Yip, in *Properties of Liquid Metals: Proceedings,* edited by S. Takeuchi and S. Takeuchi (Halsted Press, New York, 1973), p. 119.

[13] W. Glaser, S. Hagen, U. Löffler, J. B. Suck, and W. Schommers, in *Properties of Liquid Metals: Proceedings,* edited by S. Takeuchi and S. Takeuchi (Halsted Press, New York, 1973), p. 111.

[14] U. Löffler, Kernforschungslage Karlsruhe, 1973 (unpublished).

[15] P. G. de Gennes, Physica (Utrecht) 25, 825 (1959).

[16] Institut Laue-Langevin Neutron Beam Facilities Handbook, Grenoble, France, 1975 (unpublished).

[17] J. K. Kjems and P. A. Reynolds, in *Neutron Inelastic Scattering, 1972* (UNIPUB, New York, 1973), p. 733.

[18] K. Sköld, J. M. Rowe, G. Ostrowski, and P. D. Randolph, Phys. Rev. A 6, 1107 (1972).

[19] G. H. Vineyard, Phys. Rev. 110, 999 (1958).

[20] V. Heine and D. Weaire, Phys. Rev. 152, 603 (1966); V. Heine, J. Phys. C: Solid State Phys. 1, 222 (1968); J. E. Inglesfield, J. Phys. C: Solid State Phys. 1, 1337 (1968).

[21] J. P. Badiali and C. Regnaut, Phys. Status Solidi (b) 63, 555 (1974).

Magnetic Interactions in Dilute *Ag*Mn Alloys

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Measurements are presented of the magnetization M up to 50 kG between 1.8 and 4.2 K and the susceptibility χ from 1.8 to 100 K of three dilute *Ag*Mn alloys of nominal concentrations 630, 1000, and 1370 ppm Mn. Both M and χ follow scaling laws well. The strength V_0 of the Ruderman-Kittel-Kasuya-Yosida interaction between two Mn impurities has been determined from expressions for the high-field behavior of M in the presence of such interaction, yielded by a virial expansion of the free energy. The value for V_0 is $(2.4 \pm 0.4) \times 10^{-25}$ eV cm^3. The effective $|J|$ value estimated from V_0 is 0.7 ± 0.1 eV.

Recently interest in alloy research has been shifting from the extremely dilute region ($T_I < T_K$, where T_I is the measure of average interactions between solute atoms and T_K is the Kondo temperature) in which the single-impurity Kondo effect dominates to the more concentrated region in which the interactions between impurities can no longer be neglected. The strength V_0 of the Ruderman-Kittel-Kasuya-Yosida[1] (RKKY) interaction, $V(r) = V_0 \cos(2k_F r)/r^3$ (for $k_F r \gg \pi$), between two Mn impurities and the effective $|J|$ value in *Ag*Mn alloys have been determined by magnetization measurements at high fields and are presented here. The *Ag*Mn alloy system was chosen because T_K is very small, so that in the dilute limit the solute has a good magnetic moment down to very low temperatures; and because the electronic structure of Ag is simple it can be safely approximated by a free-electron model. It is thus unlike *Au*Fe, for example, where the effect of T_K on the interaction behavior is clear in the results of Laborde and Radhakrishna.[2]

Three *Ag*Mn alloys with respective nominal concentrations of 630, 1000, and 1370 ppm Mn were examined. The starting materials were 99.9999% Specpure Ag (Johnson Matthey) and 99.99%-pure Mn from Koch-Light Labs Ltd. The samples were made by diluting a 5.43-at.% *Ag*Mn master alloy in an arc furnace filled with $\frac{2}{3}$ atm argon, and forming short cylinders of about 3 mm diameter. All samples were cleaned by a mixture of NH_4OH and H_2O_2. Measurements were carried out by a force (Faraday) method using a superconducting solenoid (0–50 kG) with separate super conducting gradient field coils; the forces were measured with a Beckman LM-600 microbalance.

Temperatures T were derived from the He vapor pressure from 1.8 to 4.2 K, from the resistance of an Allen Bradley carbon resistor from 4.2 to 25 K, and at higher temperatures from the reading of a copper-Constantan thermocouple with a reference junction in liquid nitrogen.

The magnetic susceptibility χ of the Mn solute was assumed to be $\chi_{\rm alloy} - \chi_{\rm Ag}$, where $\chi_{\rm alloy}$ is the susceptibility of the alloy and $\chi_{\rm Ag}$ is that of Ag. $\chi_{\rm Ag}$ has been measured from 1.8 to 100 K, and since it changes only very slightly with temperature compared with the change of χ, the value -1.86×10^{-7} emu/g for $\chi_{\rm Ag}$ was used as the correction for the whole of this temperature range. The initial susceptibility follows a Curie-Weiss behavior and from the Curie constant the spin $S = 2.23 \pm 0.01$ per Mn atom was determined, which is in good agreement with the values of Mizuno[3] and Flouquet,[4] where in this and later calculations the assumption $g = 2$ was made.

The RKKY impurity-impurity interaction and the single-impurity Kondo effect will both reduce the magnetization M below the Brillouin function appropriate for free spins, and also inhibit the approach to saturation of M. From the RKKY interaction, scaling laws[5] $M/n = F_1(T/n, H/n)$ and $\chi = F_2(T/n)$ were derived, where H is the magnetic field and the concentration n acts as the scaling parameter. The M/n-versus-H/n behavior has been plotted in Fig. 1 for the three alloys by fixing the reduced temperature T/n at 3.0×10^{-3} K/(ppm Mn), and χ versus T/n in Fig. 2 for the 630- and 1370-ppm alloys. The experimental results follow the scaling laws well. Another characteristic feature of magnetic interaction which arises from the RKKY interaction is that the magnetization per impurity atom, at a given H and T, decreases as n increases. This characteristic feature can be seen implicitly in Fig. 3. The above evidence thus suggests that the RKKY interaction, rather than the single-impurity Kondo effect or crystal-field splitting, dominates the magnetic behavior of the alloys measured.

By using a virial expansion of the free energy in a power series in the concentration of magnetic impurities to investigate the effect of the RKKY interaction on the thermodynamic function of dilute magnetic alloys, Larkin and Khmel'nitskii[6] derived the following temperature-independent expression for magnetization M at the kT

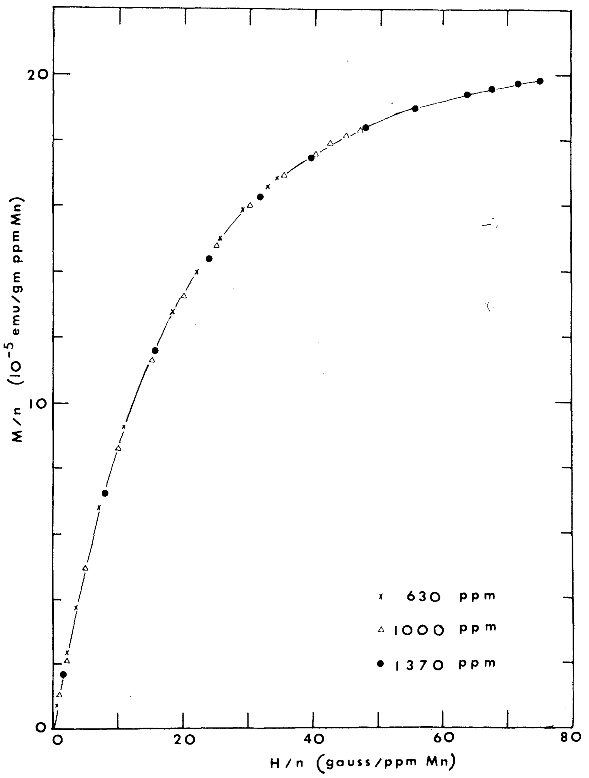

FIG. 1. Impurity magnetization M divided by concentration n as a function of magnetic field H divided by n at the fixed reduced temperature $T/n = 3.0\times10^{-3}$ K/(ppm Mn) for 630-, 1000-, and 1370-ppm-Mn *Ag*Mn alloys.

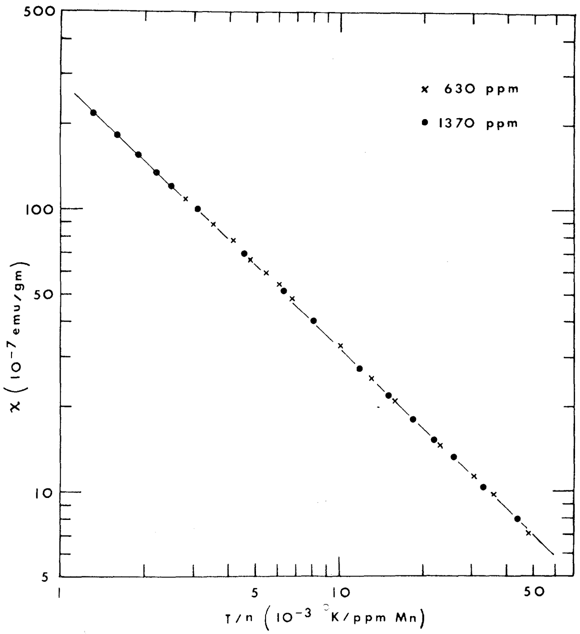

FIG. 2. Magnetic susceptibility χ as a function of reduced temperature T/n for 630- and 1370-ppm-Mn *Ag*Mn alloys.

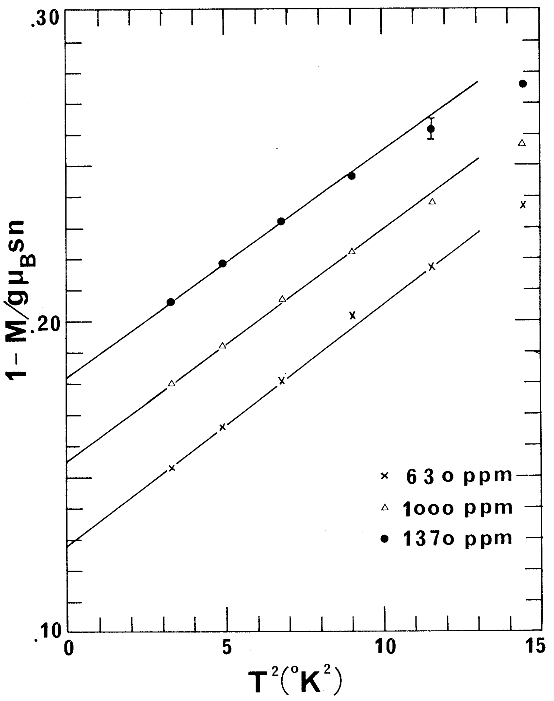

FIG. 3. Fractional deviation of magnetization M, taken at field $H = 45$ kG, from the saturation magnetization $g\mu_B Sn$. $1-M/g\mu_B Sn$ as a function of T^2 for 630-, 1000-, and 1370-ppm-Mn alloys.

$\ll \mu_B H$ limit,

$$M = g\mu_B Sn[1 - 2(2S+1)nV_0/3g\mu_B H].$$

However, under practical laboratory conditions (say $H = 45$ kG) the $kT \ll \mu_B H$ limit might be difficult to achieve and the effect of temperature should be considered as well. In practice one is dealing with a situation where the temperature cannot be regarded as very small, but provided that $8nV_0/3kT < 1$ the use of a pair approximation will still be valid. The formal considerations that apply to extensions beyond the $\mu_B H \gg kT$ limit examined by Larkin and Khmel'nitskii apply strictly only to the $S = \frac{1}{2}$ case, but the general form of the dependence of magnetization on field and temperature is likely to be similar to that derived simply by following the Larkin and Khmel'nitskii procedure and arbitrarily equating the coefficients of the temperature-independent term to yield

$$M = g\mu_B Sn\{1 - 2(2S+1)(nV_0/3g\mu_B H) \times [1 + \tfrac{1}{3}(\pi k/g\mu_B H)^2 T^2]\}$$

for the small-$kT/\mu_B H$ case.

[Matho (private communication) has derived much more rigorously an expression of just this form. This work extends previous work of Matho and Beal-Monod[7-9] which both uses the virial-expansion approach and introduces the complications involved when the Kondo effect is taken into account.]

The fractional deviation of magnetization M (taken at field $H = 45$ kG) from the saturation magnetization, under the assumption that this is $ng\mu_B S$, has been plotted as $1 - M/g\mu_B Sn$ versus T^2 in Fig. 3. A T^2 dependence is observed clearly, and a linear relation between $1 - M/g\mu_B Sn$ and n at a fixed T can also be seen implicitly. From the intercepts at $T^2 = 0$ of the straight lines, the strength of the RKKY interaction between two Mn atoms in *Ag*Mn $V_0 = (2.4 \pm 0.4) \times 10^{-25}$ eV cm^3 has been determined, where 1 ppm Mn $= 5.91 \times 10^{16}$ (Mn atoms)/cm. Since our values of $8nV_0/kT$ are thus in the range 0.1–0.3 the use of a pair approximation is justified.

The effective $|J|$ value of the s-d exchange interaction $V_{sd} = -J\vec{S}\cdot\vec{s}$ between the spin operators S and s for the spin momenta of Mn and conduction electron of Ag can be estimated by putting the values of V_0 and S obtained above into the equation[10]

$$V_0/2 = 9\pi^2 J^2 S(S+1)/64\sqrt{2}E_F k_F^3,$$

where the factor $\frac{1}{2}$ on the left-hand side appears because V_0 is the strength defined between two impurities, with the free-electron parameters $E_F = 5.48$ eV and $k_F = 1.2 \times 10^8$ cm^{-1}. The effective $|J|$ value of 0.7 ± 1 eV has been determined, which is between on the one hand the value of 1.1 eV obtained by Mizuno[3] from NMR studies and on the other hand the value of 0.22 eV derived by Flouquet[4] from γ-ray anisotropy measurements and that of 0.26 eV yielded[8] by an analysis of the resistivity data of Jha and Jericho.[11]

It must be emphasized that comparisons of estimates of $|J|$ from measurements of different types must be treated with caution to avoid pitfalls long recognized in paramagnetic-resonance studies of alloys.

We thank P. L. Li for the preliminary discussions of the curvature of magnetization curves of other alloys of ours, which led to our wish to make further investigation into the effects of impurity interaction. We are grateful for stimulating and helpful discussions with J. C. Liu, K. Matho, and N. Rivier. We also thank F. W. Smith, A. E. Bell, and R. Aoki for their interests and H. E. N. Stone for assistance with sample

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[1] M. A. Ruderman and C. Kittel, Phys. Rev. 96, 99 (1954); R. Kasuya, Prog. Theor. Phys. 16, 45 (1956); K. Yosida, Phys. Rev. 106, 893 (1957).

[2] O. Laborde and P. Radhakrishna, Solid State Commun. 9, 701 (1971).

[3] K. Mizuno, J. Phys. Soc. Jpn. 30, 742 (1971).

[4] J. Flouquet, Phys. Rev. Lett. 25, 288 (1970).

[5] J. Souletie and R. Tournier, J. Low Temp. Phys. 1, 95 (1969).

[6] A. I. Larkin and D. E. Khmel'nitskii, Zh. Eksp. Teor. Fiz. 58, 1789 (1970) [Sov. Phys. JETP 31, 958 (1970)].

[7] K. Matho and M.-T. Beal-Monod, J. Phys. (Paris), Colloq. 32, C1-213 (1971).

[8] K. Matho and M.-T. Beal-Monod, Phys. Rev. B 5, 1899 (1972).

[9] K. Matho and M.-T. Beal-Monod, J. Phys. F: Metal Phys. 4, 848 (1974).

[10] See, for example, W. A. Harrison, *Solid State Theory* (McGraw-Hill, New York, 1970), Chaps. IV and V.

[11] D. Jha and M. H. Jericho, Phys. Rev. B 3, 147 (1971).

Thermomodulation Study of Plasmons and Longitudinal Excitons in Alkali Halides*

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Thermoreflectance measurements on LiF and KCl were made from 10 to 30 eV using synchrotron radiation. Analysis of the differential dielectric and electron-energy-loss functions shows that the energy-loss peak generally attributed to the valence plasmon actually arises from the plasmon and overlapping longitudinal-exciton-like peak(s).

Different interpretations have been proposed for the strong peak in the electron-energy-loss spectra on the high-energy side of the fundamental absorption edge of alkali halides. The first was to associate it with the plasma resonance of the valence electrons.[1-3] The dielectric function, $\tilde{\epsilon}=\epsilon_1+i\epsilon_2$, has the behavior characteristic of a free-electron gas: ϵ_1 crosses zero with positive slope and ϵ_2 is small, decreasing with increasing energy. The energy of the loss peak is higher than the free-electron plasmon energy, $h\omega_p$, calculated for 6 electrons per molecule. This shift is due to the presence of interband transitions at lower energies.[4] Sueoka[5] measured the angular dispersion of the loss peak in LiF (25.3 eV) and found a strong plasmonlike dispersion. A totally different dispersion was found by Creuzburg and Raether,[6] who found essentially no momentum dependence, as expected for single-particle excitations. They concluded that the loss peak is determined by one-electron transitions. A third interpretation considered both the peak in ϵ_2 and that in $\mathrm{Im}(-1/\tilde{\epsilon})$ as arising from the simultaneous creation of two excitons.[7,8] As expected, the peak in ϵ_2 occurs at an energy approximately twice that of the first exciton peak. The calculated shift of the loss peak toward higher energies is in agreement with experiment. Gout and Pradal[9] found the 25.3-eV loss peak actually had two components. Antinori, Balzarotti, and Piacentini[10] noticed that the structure in $\mathrm{Im}(-1/\tilde{\epsilon})$ is connected with the one in ϵ_2. They found for KBr that the energy-loss function calculated by fitting the ϵ_2 peak with a Lorentzian oscillator peaks at the same energy as the experimental one. In several alkali halides the peak in ϵ_2 shows a multiplet structure as does the loss peak. In such cases the high-energy component of the loss structure has been associated with the plasma resonance and the other with interband transitions.[9] LiF is unique, for ϵ_2 shows a single feature at 23 eV[11] while the energy-loss structure is double.[9]

No additional experimental data exist so far to test these interpretations. Since most of the alkali halides show these features, they probably have a common origin. We measured the thermoreflectance (TR) of several alkali halides between 12 and 30 eV, where these structures are present. By means of Kramers-Kronig analysis, we obtained the thermomodulation (TM) differential optical constants $\Delta\tilde{\epsilon}=\Delta\epsilon_1+i\Delta\epsilon_2$, and $\Delta[\mathrm{Im}(-1/\tilde{\epsilon})]$ the differential electron-energy-loss function (ΔEEL).

The experimental technique[12] will be described in detail elsewhere. Synchroton radiation emitted