

a shift nor a broadening of the emitted line with respect to the incident line has been observed within the limits of error of $\pm 3 \text{ \AA}$ given by the resolution of the monochromator. Furthermore, the intensity of emission obtained by irradiating the foil with light of a narrow spectral width has been found to be the same as that obtained at the same frequency by irradiating the foil with a large frequency band ($>1000 \text{ \AA}$). These results are in good agreement with the emission experiments made on silver.⁴ Obviously, the plasma behaves like a resonator.

The half-width of the emitted line is 400 \AA or 460 meV in good agreement with the energy-loss measurements. (However, these two values are not directly to be compared, since the width of the emitted line is dependent on the observation angle θ . It needs to be extrapolated to $\theta=0$ in order to be compared with the half-width of the electron energy loss. For this purpose, the half-width has to be measured

as a function of θ . These experiments are in preparation. This correction, however, is not of great importance.)

It may be added that the maximum of the emission can be found at longer wavelengths than 3300 \AA , depending on the conditions of evaporation; the minimum of the transmission (plasma-resonance absorption) is displaced in the same way.

These experiments show that the plasma of free electrons, excited by light, also shows the phenomenon of plasma-resonance emission.

¹J. Brambring and H. Raether, Phys. Rev. Letters 15, 23, 882 (1965); Z. Physik 199, 118 (1967).
W. Steinmann, J. Hofmann, and K. Stettmaier, Phys. Letters 23, 234 (1966).

²J. Brambring and H. Raether, Z. Naturforsch 21a, 9, 1527 (1966).

³C. Kunz, Z. Physik 196, 311 (1966).

⁴P. Schreiber and H. Raether, Z. Naturforsch 21a, 2116 (1966).

OBSERVATION OF NAGAOKA'S BOUND STATE FOR CONDUCTION ELECTRONS IN DILUTE MAGNETIC ALLOYS*

M. D. Daybell† and W. A. Steyert

Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico

(Received 3 February 1967)

Recent theories differ as to whether a negative s - d exchange interaction between a localized-impurity moment in a dilute alloy and the conduction-electron spins will result in the formation below some critical temperature T_C of a quasibound state analogous to the Cooper pair in the theory of superconductivity,¹⁻³ or will merely cause a strongly temperature-dependent scattering of conduction electrons at low temperatures.⁴

Electrical resistivity measurements over the three-decade temperature range from 40 mdeg to 40°K reported here for dilute alloys of iron in copper permit a clear choice between the two predictions. Preliminary susceptibility measurements on the same material also tend to support the bound-state model. Quantitative agreement below T_C between Nagaoka's theory and the resistivity data permit T_C and hence Kondo's exchange coupling constant J to be determined. Above T_C serious disagreement is found with the predictions of pertur-

bation theory, heretofore considered by most authors to be valid in the region sufficiently far above this temperature. The high value of T_C (16°K) and the observed absence of impurity-impurity interactions make dilute Cu-Fe a nearly ideal system for studying the quantitative features of the low-temperature state.

The alloy samples were vacuum cast in alumina-coated spherical graphite molds 11 mm in diameter, using high-purity copper combined with a suitable quantity of a Cu-0.5%Fe master alloy to create the very dilute samples needed. The larger susceptibility sample was prepared from the same master in a similar mold of different shape. All samples were chemically etched before use. The resistivity ratio at room temperature to helium temperature of a pure-copper sample prepared using the same techniques was greater than 900. Absence of a resistivity minimum in this sample indicates that it contained less than 2 ppm iron. The samples were supported below a He³-He⁴

dilution refrigerator mounted in a glass vacuum space immersed in liquid helium.

An ac mutual-inductance eddy-current bridge specifically designed for this work was used to measure resistivity and susceptibility, and is to be described elsewhere.⁵ The eddy-current method measures the average bulk resistivity, and is insensitive to possible resistivity variations caused by oxidation of the impurity atoms near the sample surface. Resistivity measurements could be made to an absolute accuracy of better than 3%. Dissipation in the samples was typically a few nanowatts. Susceptibility changes of 10^{-8} emu/g were detectable, although reproducibility was worse than this because of small thermal effects in the pickup coils. Eddy-current effects were eliminated by laminating the susceptibility sample.

The resistivity increment per atomic part per million (or "ppm") iron impurity introduced was obtained by subtracting the resistivity of the pure copper sample from that of the various alloys and dividing by the impurity concentration (Fig. 1).⁶ White's earlier data⁷ for a much more concentrated alloy are included in the figure to support the conclusion that cooperative effects are absent in the range of concentrations under study. His data were presented with an arbitrary vertical scale factor which

we have chosen in order to make his sub-1°K data agree with ours.

The resistivity predicted by Nagaoka's low-temperature solution for his model, which results in the formation of a quasibound state below T_C , can be seen to agree quite well with the measured values if T_C is taken to be 16°K, as shown.⁸

The temperature dependence of the change in the paramagnetic susceptibility of a 110-ppm Fe-Cu alloy at low temperatures is shown in Fig. 2. Part of the more accurate higher-temperature data of Hurd⁹ above 5.6°K is also shown. A marked decrease in the effective magnetic moment is evident in the region just below T_C , although the exact behavior of the susceptibility below the critical temperature may deviate from that of Fig. 2. The susceptibility change of the sample with temperature is extremely small (less than 1/3000 that of cerium magnesium nitrate), and many other materials (copper oxide, for example) are quite magnetic in this region. Further measurements of this effect are planned. Nagaoka predicts a significant reduction in the effective moment as the bound state is formed, but it is not as large as the observed decrease. The possibility that the effective moment may actually vanish as T goes to 0 as predicted by other bound-state

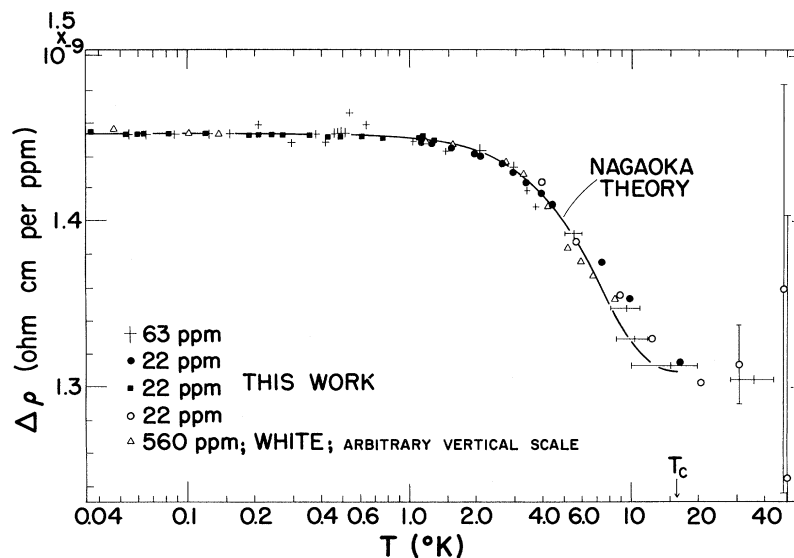


FIG. 1. Impurity contribution to the electrical resistivity per at. ppm iron impurity in copper. Except where error bars appear, errors on open circles and crosses are ± 0.01 n Ω cm/ppm, errors on filled symbols are 0.001 n Ω cm/ppm, and temperature errors are negligible. Absolute error in the resistivity per ppm is approximately 10%, resulting primarily from uncertainties in the impurity concentration. Large error bars at right result from uncertainties in the appropriate host resistivity to subtract for one run made without a pure Cu reference sample.

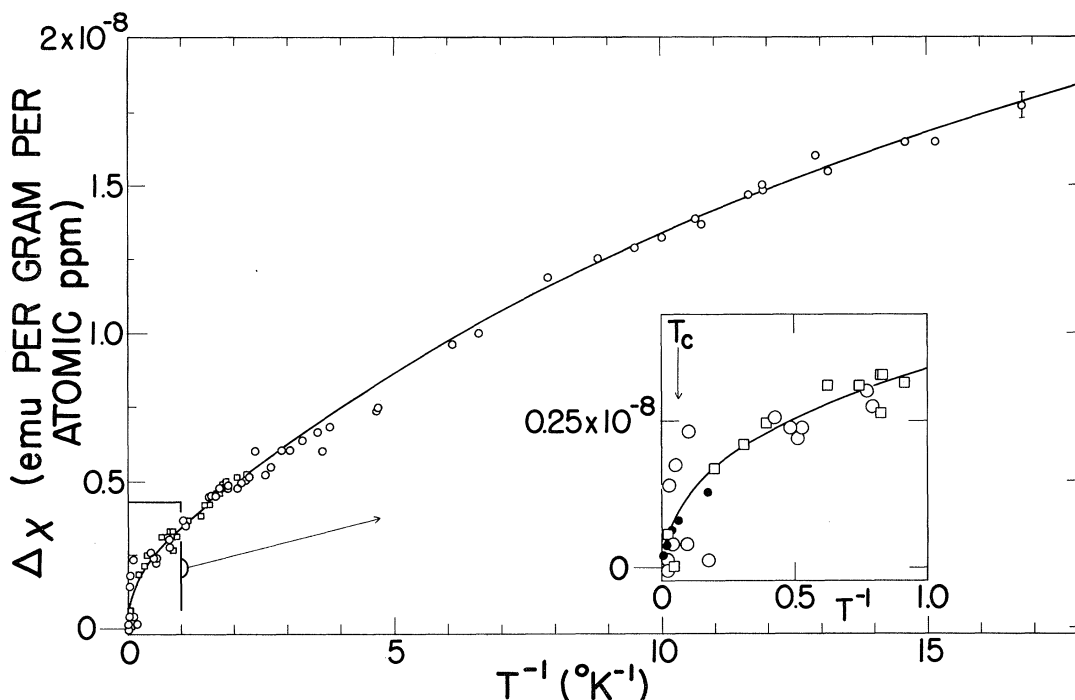


FIG. 2. Incremental susceptibility per gram of alloy per ppm versus reciprocal temperature for 110-ppm iron impurity in copper. The open circles and squares represent two separate runs. Data above 5.6°K for a 9.0-ppm alloy, given by Hurd, are compared (solid circles) with the present data in the expanded insert. $\Delta\chi$ is accurate to within 10%. The solid curve is the function $A/(T+T_A)+B/(T+T_B)$, where from Hurd's data, $A=2.52\times 10^{-8}$ emu/g ppm, $T_A=14^{\circ}\text{K}$; and from the present data, $B=0.168\times 10^{-8}$ emu/g ppm, $T_B=0.045^{\circ}\text{K}$. In effective Bohr magnetons per iron atom, A corresponds to $3.6\mu_B$, B to $0.92\mu_B$.

calculations² cannot be excluded.

It is not possible to make a comparison between theory and experiment in the problem of the s - d interaction by examining the resistivity, the susceptibility, and the specific heat.¹⁰ Below 6°K there is excellent agreement between the measured resistivity and Nagaoka's quasibound-state theory. Moderately good agreement with the low-temperature form of this theory is obtained right up to T_c . The present data show that predictions by Kondo, Abrikosov, Suhl,⁴ and others that there would be a peak in the anomalous low-temperature resistivity of these systems are not fulfilled. These predictions did not allow for the formation of a bound state.

Many authors, including Nagaoka, have made perturbation calculations of $\Delta\rho$ and $\Delta\chi$ that diverge logarithmically with T near T_c but break down below T_c . None of the predicted anomalous behavior is observed. A predicted zero in $\Delta\chi$ near $2T_c$ is also absent. It is possible that lattice vibrations quench the interaction mechanism at these higher temperatures or

that the resistivity of the pure metal and the impurity are not simply additive.

The total entropy change of $R \ln 2$ (per mole of impurity) contained in the specific-heat anomaly observed in these systems¹⁰ below about 15°K is somewhat smaller than Nagaoka would predict¹¹ (R is the gas constant per mole). His low-temperature limit for ΔC_v , however, agrees with the measured temperature dependence in the range 0.4 to 1.5°K if T_c is taken as 16°K .

We may conclude from these results that below the critical temperature the features of the quasibound-state model put forth by Nagaoka are essentially correct.

We would like to thank Mr. Don Hull for help with sample preparation, and Dr. Martin Maley for valuable discussions.

*Work performed under the auspices of the U. S. Atomic Energy Commission.

†Associated Rocky Mountain Universities visiting scientist on leave from New Mexico State University.

¹Y. Nagaoka, Phys. Rev. **138**, A1112 (1965).

²F. Takano and T. Ogawa, *Progr. Theoret. Phys.* (Kyoto) **35**, 343 (1966).

³K. Yosida, *Phys. Rev.* **147**, 223 (1966).

⁴J. Kondo, *Progr. Theoret. Phys.* (Kyoto) **34**, 204 (1965); A. A. Abrikosov, *Physics* **2**, 5 (1965); H. Suhl, *Physics* **2**, 39 (1965).

⁵Melvin D. Daybell, to be published.

⁶C. A. Domenicali and E. L. Christenson, *J. Appl. Phys.* **32**, 2450 (1961), show changes of about 0.01 nΩ cm/ppm between 30 and 80°K for their 500- to 3000-ppm Cu(Fe) alloys.

⁷G. White, *Can. J. Phys.* **33**, 119 (1955).

⁸The temperature-independent resistivity arising from ordinary scattering by the impurity atoms has been taken as 1.31 nΩ cm/ppm. This and T_c are the

only fitting parameters used. Other constants appearing in Nagaoka's resistivity expression [Eq. (5.10), Ref. 1] are the effective mass m^* of the copper conduction electrons, and the average density of states per atom $\bar{\rho}/N$ in the copper conduction band [cf. Eq. (3.9), Ref. 1]. We take $m^* = 1.4 m_e$, and, using a mean bandwidth $2D = 10$ eV, $\bar{\rho}/N$ is 0.2/eV per atom, using the fact that there are two conduction band states per atom. With $T_c = 16^\circ\text{K}$, $(J\bar{\rho}/N)^{-1}$ is found to be 8.32, making $J = 0.6$ eV.

⁹C. M. Hurd, *Cryogen.* **6**, 264 (1966).

¹⁰J. P. Franck, F. D. Manchester, and D. L. Martin, *Proc. Roy. Soc. (London)* **A263**, 494 (1961); F. Du Chatenier and J. De Nobel, *Physica* **32**, 1097 (1966).

¹¹Y. Nagaoka, to be published.

ISOTOPES OF ELEMENT 102 WITH MASS 251 TO 258†

Albert Ghiorso, Torbjorn Sikkeland, and Matti J. Nurmi*

Lawrence Radiation Laboratory, University of California, Berkeley, California

(Received 9 February 1967)

The purpose of this Letter is to outline recent information obtained in our laboratory concerning a wide range of isotopes of the element with atomic number 102. A more detailed article is presently being prepared for submission elsewhere.

We have used ¹²C and ¹³C ions accelerated by the Berkeley heavy-ion linear accelerator (HILAC) to bombard essentially monoisotopic targets of ²⁴⁴Cm, ²⁴⁶Cm, and ²⁴⁸Cm for the production of the isotopes ²⁵¹102 to ²⁵⁸102. The apparatus is an elaboration of a simple principle first observed in this laboratory some years ago.¹ Atoms recoiling from the target are stopped in a stream of helium at 600 Torr and carried by this gas through an orifice about 0.2 mm in diameter into an evacuated space. The gas jet impinges a few millimeters away on the periphery of a wheel and a large fraction (~80%) of the heavy atoms attach themselves to its surface. At regular intervals the wheel is digitally rotated about 50° to expose the collected atoms to Au-Si surface-barrier alpha-particle detectors. In this series of experiments four detectors, equally spaced along the circumference of the wheel, were used simultaneously in order to obtain half-life information as well as alpha-particle energies. The targets, made by molecular deposition, were 0.2 to 0.5 mg/cm² of curium oxide on 4 to 5 mg/cm² beryllium metal. The carbon-ion beam currents used were typically 2×10^{12} particles/

sec in an area of 0.2 cm². Changes in bombarding energy were made by inserting different thicknesses of Be degrader foils in the beam path so that excitation functions for the alpha-particle activities could be determined.

The electronic circuitry to analyze the pulse outputs from the individual detectors was conventional. After preamplification with charge-sensitive amplifiers located near the detectors, the pulses were shaped by delay lines to one microsecond and further amplified in the counting area. They were then sorted with a two-parameter analyzer into four two-hundred-channel groups. The resolution of the system varied from 25 to 50 keV full width at half-maximum depending on the detector used. Spontaneous fissions were recorded by discriminators set to trigger on pulses greater than 30 MeV in amplitude. It was necessary to gate the system off during each beam pulse to prevent spurious signals from neutron reactions and thus a 20% loss was suffered. The total counting efficiency, defined as the ratio of the counts observed to the alpha disintegrations undergone by the nuclei transmuted from the target, was about 10%. Supplementary measurements of spontaneous fission activity were made in many experiments with the aid of mica detectors.²

Fortunately the production cross sections to form the element-102 isotopes by these reactions are in the range from 10^{-31} to 10^{-30} cm² so that it has been possible to make mea-