Resonant free-induction signals in metals in high vacuum

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An emission signal has been observed in several metals after an intense microwave pulse. The signal has a resonance in the magnetic field with a decay time $\sim 10^{-4}$ s. The effect apparently results from the appearance of electron surface states on metal samples in high vacuum.

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Important among the various physical methods used for studying metals are the rf-spectroscopic methods which make use of effects such as cyclotron resonance[1] and the paramagnetic resonance involving conduction electrons and localized magnetic moments. We report here the use of a new method for studying metals. This method makes use of the resonant emission in the form of an induction signal in a magnetic field perpendicular to the surface of the sample.

Measurements were taken from flat metal samples ranging in thickness from a few microns to 5 mm. The glass ampules holding the metal samples were pumped down by a roughing pump to 10^{-2} Torr and sealed off. Since the experiments were carried out at 2 and 4.2 K, the vacuum in the sealed-off ampules reached 10^{-7} Torr. The ampules, with the samples, were placed in a cylindrical resonator and bombarded with microwave pulses 4×10^{-8} s long at a frequency of 9.4 GHz. The microwave pulse was followed by the appearance of an induction signal lasting 10^{-4} s (Fig. 1). The power level of this response varied over the range 10^{-8}–10^{-9} W for the various samples. The sensitivity of the detection was 10^{-12} W. Interestingly, the emission following the microwave pulse appears only at a certain strictly defined magnetic field, and it is clearly of a resonant nature. The signal is observed only in the sealed-off ampules; if the seal is broken, the signal disappears completely. The induction is also erased if paraffin is poured over the sample in a vacuum or if the ampule is filled with gaseous helium. Furthermore, if the sample is placed directly in liquid helium, the signal is not observed. Irregularities with sizes up to 0.1 mm deliberately produced on the polished metal surfaces did not have any significant effect on the induction signal. Annealing

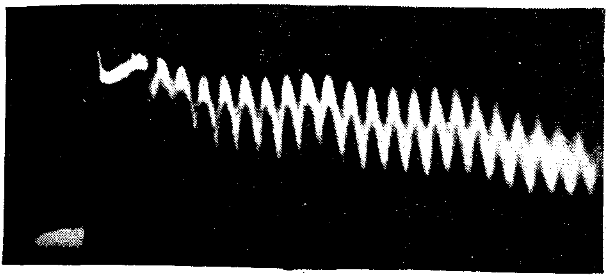

FIG. 1. Oscilloscope trace of the free-induction signal for an Al sample. The induction decay is amplitude-modulated at 4×10^5 Hz.

the copper sample in vacuum (10^{-5} Torr) for a day in a quartz ampule at 950 °C reduced the signal by an order of magnitude. A subsequent chemical polishing of the surface of the annealed sample restored the original signal intensity. Cryogenic pumping with a simultaneous annealing of the sample was used instead of the roughing pump to rule out the possibility that vapor of the vacuum oil could reach the samples. The same results were obtained.

For a detailed study of the dependence of the emission intensity on the magnetic field, we strobed the induction signal and sent it to an integrator and then to the "Y" input of an X, Y chart recorder. To the "X" input we sent a voltage proportional to the linearly varying magnetic field. Figure 2 shows the shape of the emission line for a copper sample prepared by rolling a single crystal. The line peaks correspond to g-factors of 2.004 and 1.981. Similar signals have been observed for a long list of metals (Be, Mg, Al, Fe, Co, Ni, Ag, Au, Cd, In, Sn, Pb, Cu, Hg, Ce, Sm, Eu, Tb, Dy, La, Ho, Er). Each metal has its own distinctive line widths, intensities, and positions along the field scale. The most intense line is exhibited by beryllium. None of the lines had a

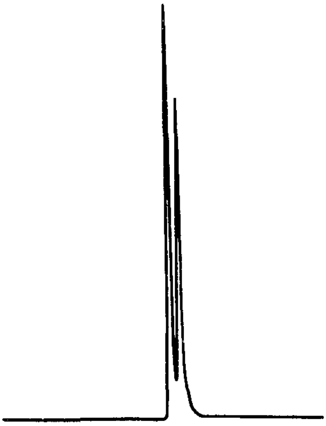

FIG. 2. Shape of the emission line for a Cu sample. The line peaks correspond to fields of 3344 and 3382 Oe. The widths of the lines along the field scale are 7 and 8 Oe, respectively. The modulated frequency of the microwave pulses is 9381.4 MHz.

width exceeding 10–15 Oe. The greatest deviation from $g = 2$ was observed for iron and terbium. The iron samples exhibit two lines, with $g = 1.909$ and 1.872; the terbium also exhibits two lines, with $g = 2.161$ and 1.899. It was established in the course of these experiments that the effect is observed only if a static magnetic field is applied perpendicular to the surface of the sample; a deviation of H by a few degrees from the perpendicular orientation caused the signal to disappear. A sample cut from copper foil and a copper single crystal grown especially for the purpose did not differ in terms of the position of the line along the field scale, the line intensity, or the line shape. The nature of the observed decay is reminiscent of the well-known excitation of sound in a metal by an intense microwave pulse.[2,3] However, the fact that the induction appears in a resonant manner as a function of the magnetic field, the slow decay, and the independence of the effect from the sample thickness all make this assumption improbable. Experiments carried out with copper samples containing the paramagnetic impurities Mn and Er yielded results agreeing with those for pure copper; the implication is that the observed effect is of a surface nature. We might note that the same copper samples with the Er impurity produce an ESR signal with a g-factor ~ 6.6 both in high vacuum and in ampules whose hermetic seal has been broken.

These results, combined with the fact that the signal disappears when the metal samples make contact with paraffin, air, or helium, suggest that the observed effect results from a cyclotron resonance involving surface electrons. Tamm[4] describes the electron energy spectrum at a crystal-vacuum interface in the direction perpendicular to the surface as discrete, i.e., quantized. Such electrons on a surface may be regarded as two-dimensional. In a magnetic field oriented perpendicular to the surface, the two-dimensional motion of free electrons transforms into a set of quantized Landau levels with a splitting $eH\hbar/mc$, where m is the effective mass. The actual states are of course chemisorption states because of the interaction of the metal surface with the foreigh gas atoms. Qualitatively, however, the behavior of the electrons in the surface states is the same.[5] As the temperature is lowered, surfaces of this type undergo a "restructuring" in connection with a change in the symmetry of the surface unit cell.[6] This effect may give rise to different cyclotron masses and to the appearance of several lines, as in Fig. 2.

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