Multiple Magnon Excitation in NiO by Electron Tunneling

D. C. Tsui, R. E. Dietz, and L. R. Walker
Bell Telephone Laboratories, Murray Hill, New Jersey 07974
(Received 8 November 1971)

We report our observation of magnon excitation in antiferromagnetic NiO by electron tunneling through single-crystal Ni-NiO-Pb junctions. We compare the derivative of the tunnel conductance of the junction with a realistic magnon density of states of NiO and show that the observed structures indeed reflect the one-magnon, two-magnon, and three-magnon density of states of NiO.

Numerous experiments have shown that tunneling electrons can excite phonons of the insulating barrier in a tunnel junction.[1] Although similar excitation of magnons in the barrier has been anticipated,[2] no convincing experimental results have yet been reported. Thompson, Holtzberg, and von Molnar[3] observed a zero-bias resistance peak in degenerate EuS-In junctions, and tentatively attributed it to electron tunneling with magnon emission rather than to a two-step impurity-assisted tunneling.[4]

In this Letter, we wish to report our observation of magnon excitation in antiferromagnetic NiO by electron tunneling through single-crystal Ni-NiO-Pb junctions. We compare the derivative of the tunnel conductance (d^2I/dV^2) of the junction with a realistic magnon density of states of NiO and show that the prominent structures observed in the curves of d^2I/dV^2 versus bias (V) indeed reflect the one-magnon (1M), two-magnon (2M), and three-magnon (3M) density of states of NiO. We also discuss some unexplained electron tunneling data in the literature[5,6] which we now identify as due to magnon excitation in NiO.

The tunnel junctions are prepared by thermally growing an oxide layer on a freshly cleaned single-crystal Ni surface. The surface is first electropolished and then cleaned by low-energy ion bombardment in a vacuum of about 2×10^{-4} Torr argon pressure. The oxidation takes place in air at 360°C for two hours. The sample surface is then insulated with collodion except for a narrow strip in the center, and cross strips of Pb about 2000 Å thick are evaporated on the surface in a vacuum of 2×10^{-6} Torr.

The junction I-V characteristic and its first and second derivatives are measured using standard techniques.[7] The results presented in this Letter are obtained by recording the data for dynamic resistance (dV/dI) versus V digitally and numerically computing the tunnel conductance $G\equiv dI/dV$ and its derivative dG/dV. We have chosen Pb films as counter-electrodes of our junctions so that the well-known tunneling characteristics of superconducting Pb[7] can be used to check if electron tunneling is indeed the dominant current-carrying mechanism through the junctions.

Figure 1 shows the curve for tunnel conductance versus bias of a Ni-NiO-Pb (normal-state) junction at 1°K. The junction is fabricated on a Ni (110) surface, and the Pb superconductivity is quenched by an applied magnetic field $H=2.5$ kG. Our interest here is in the fine structures of this curve, which have their origin in inelastic tunneling with emission of magnons of NiO and phonons of NiO, Ni, and Pb. However, before we discuss these fine structures in any detail, two comments should be made concerning the overall conductance curve. First, the conductance at low bias, $|V|\lesssim 150$ mV, has a parabolic dependence on V and shows no zero-bias anomaly of any kind. In fact, our second-derivative data show that there is no discontinuity in the slope of this curve at zero bias. Second, the rate of conductance increase with increasing bias for Ni-NiO junctions is much faster than that observed in more widely

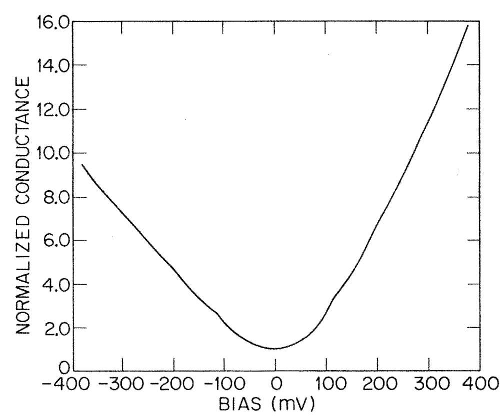

FIG. 1. Curve of normalized tunnel conductance $(dI/dV)/(dI/dV)_{V=0}$ versus bias of a Ni-NiO-Pb junction at 1°K. The junction is fabricated on a Ni (110) surface, and the Pb superconductivity is quenched by applying a magnetic field $H\approx 2.5$ kG.

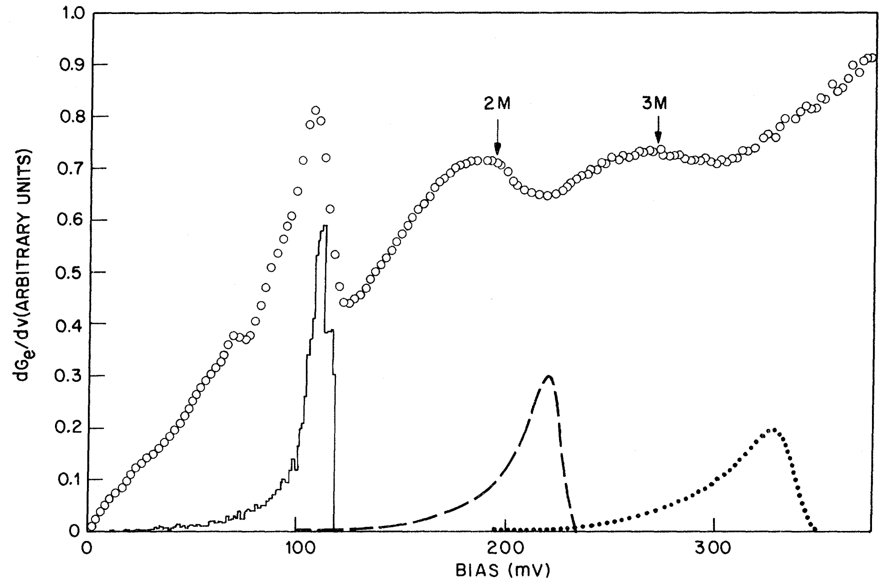

FIG. 2. The derivative of the even part of the conductance curve of Fig. 1. The open circles are the experimental data. The distribution drawn with a solid line at the bottom of the figure is the one-magnon density of states computed using $J_2 = 19.0$ meV, $J_1 = 1.36$ meV, and neglecting anisotropy. The dashed curve gives the 2M noninteracting density of states which is obtained by convoluting the computed 1M distribution with itself. The dotted curve is the 3M noninteracting density, computed by convoluting the 2M with the 1M. The arrow labeled 2M denotes the peak position of the interacting 2M Γ_3^+ spectral density observed in Raman scattering. The arrow labeled 3M likewise denotes the peak position of the interacting 3M spectral density estimated by linearly interpolating between the observed 2M and 4M Raman peaks.

studied simple metal- (Pb, Sn, Al) oxide junctions. For example, in Al-oxide-Pb junctions it usually requires a bias of approximately 400 mV to double the zero-bias conductance. As seen in Fig. 1, the background conductance due to elastic tunneling through Ni-NiO junctions increases approximately tenfold as the bias is increased to 400 mV. This strong dependence on bias is indicative of the NiO barrier being considerably lower than that of the simple metal oxides.

Previous studies of barrier phonon excitation[1] and recent theoretical-model calculations[2,8] have shown that inelastic tunneling through barrier excitation makes a contribution to the tunnel conductance which is even with respect to bias polarity. Therefore, it is convenient to separate the conductance into an even part, $G_e \equiv \frac{1}{2}[G(V) + G(-V)]$, and an odd part, $G_0 \equiv \frac{1}{2}[G(V) - G(-V)]$. The derivative of G_e gives a measure of the barrier excitation density of states superposed on a background which is the even conductance arising from elastic tunneling. Figure 2 shows the derivative of this even part of the conductance curve of Fig. 1. Although it is not clear at present what the elastic-tunneling background should be, it is nevertheless obvious that there is a series of peaks reflecting structures in the barrier-excitation density of states. The series of weak peaks near zero bias are the phonon peaks of Pb (10 mV), Ni (25 and 33 mV),[5,9] and NiO (48 and 68 mV).[10] We attribute the most prominant structure, which is a peak at $V = 107$ mV and a cutoff at $V = 122$ mV, to the magnon density of states of antiferromagnetic NiO.

The Néel temperature of NiO is $T_N = 523$°K. Above T_N it has NaCl structure, and below T_N a small rhombohedral distortion occurs. Its magnon dispersion relation can be expressed in terms of the following parameters[11]: J_2, J_1^+, J_1^-, D_1, and D_2. J_2 is the next-nearest-neighbor exchange integral which couples each Ni ion to its six neighbors on the alternate magnetic sublattice. J_1^+ and J_1 are the nearest-neighbor exchange integrals which couple the Ni ion to its six nearest neighbors of parallel spin and of antiparallel spin, respectively. D_1 and D_2 are anisotropy parameters. The values of these parameters have been determined recently from inelastic neutron scattering[11] and from Raman scattering[10] experiments. The solid curve at the bottom of Fig. 2 shows the magnon density of states of NiO computed from its magnon dispersion relation using $J_2 = 19.0$ meV, $J_1^+ = J_1^- = 1.36$ meV, and $D_1 = D_2 = 0$. A comparison of the experimental data with this magnon density of states shows conclusively that the prominent structure in the tunneling data is due to magnon excitation in NiO.

We have obtained similar results from all eight tunnel junctions (fabricated on three Ni samples) which we have studied. The energy position of the magnon peak varies from $V = 104$ to 107 mV; the energy position of the magnon cutoff varies from $V = 119$ to 122 mV. The excellent agreement of the observed cutoff energy with that of bulk NiO (118 meV) is in accord with the magnon density of states near its cutoff being due to short-wavelength magnons at the Brillouin zone edge and, therefore, being relatively insensitive to the thickness of our NiO film (~50 Å). The tunneling data also show a weak peak at $V \sim 87$ mV, which is not present in the bulk magnon density of states. This peak cannot arise from NiO phonons whose cutoff is at about 70 meV. We tentatively attribute it to the excitation of surface magnons of the NiO barrier.

We identify the two broad peaks at $V \approx 190$ and 270 mV in Fig. 2 as arising from the emission of two and three magnons, respectively, by the tunneling electrons in the NiO barrier. Since the maxima of these peaks are displaced to lower energy than those of the once- and twice-convoluted single-magnon density of states, considerable magnon-magnon interaction must occur. The observed position of the 2M peak (186 mV) agrees closely with that found by Dietz, Parisot, and Meixner[10] for the 2M Raman spectrum of NiO, namely, 194 meV. 3M processes have not been detected elsewhere, but linear interpolation between the 2M value mentioned above and that for 4M Raman scattering (349 meV)[12] would give 270 meV in agreement with the 3M maximum observed here.

The absence in the experimental data of any trace of peaks corresponding to the production of noninteracting magnons implies that this must be a rare event. On the other hand, a strong interaction must take place through the agency of J_2, since J_1 is small. The interacting magnons must then lie on one of the four simple-cubic lattices[12] into which the NiO face-centered cubic structure breaks up when J_1 is ignored. It seems reasonable to suppose that the tunneling electron fills an e_g hole on the Ni^{2+} and interacts principally with spins on the same sc lattice.

It is clear from the tunneling data that the electron-magnon coupling is strong, with about two magnons on the average dressing the electron. On the other hand, the observed NiO phonon peaks are much weaker, and the emission of multiple phonons has not been observed. Thus, the electron-phonon interaction in NiO must be weak in comparison with the electron-magnon interaction. Therefore, it is evident that the tunneling electron in NiO can be pictured more as a magnetic polaron, consisting of the electron and a spin polarization around it, than as a lattice polaron.

Finally, we discuss some unexplained tunneling data in the literature, which are related to our experiment. Adler and Chen[5] recently reported tunneling data on evaporated Ni-film–NiO-Pb junctions in the bias range $|V| \lesssim 150$ mV. They prepared their NiO junctions by either a glow discharge method or thermal oxidation. The data obtained from both types of junctions show an anomalous d^2I/dV^2 structure at $V \approx 100$ mV. The d^2I/dV^2 curve from the first type of junction shows a broad peak at $V \approx 100$ mV, and that from the second type shows a sharper peak at $V \approx 105$ mV. It is clear from our earlier discussions that the d^2I/dV^2 structure from both types of junctions arises from magnon excitation in the NiO barrier. The difference in peak energy can be explained by broadening effects on the asymmetric peak of the magnon density of states. Earlier, Rowell[6] studied Al-oxide-Ni junctions and observed a conductance structure at $V \approx \pm 120$ mV. The structure shows an abrupt increase in conductance (corresponding to a d^2I/dV^2 peak) at $V \approx \pm 110$ mV. His junctions were prepared by evaporating Ni films on oxidized Al films. Consequently, a thin layer of NiO could be formed at the oxide-Ni interface. We believe that the structure is due to magnon excitation in this thin layer of antiferromagnetic NiO.

We are most grateful to W. F. Brinkman and J. M. Rowell for many helpful discussions, and to G. E. Becker for Ni crystals, and to R. C. Fulton for computational aid.

[1]See, for example, J. M. Rowell, W. L. McMillan, and W. L. Feldman, Bull. Amer. Phys. Soc. 12, 77 (1967), and Phys. Rev. 180, 658 (1969); I. Giaever and H. R. Zeller, Phys. Rev. Lett. 21, 1385 (1968); J. G. Adler, Solid State Commun. 7, 1635 (1969).

[2]A. J. Bennet, C. B. Duke, and S. D. Silverstein, Phys. Rev. 176, 969 (1968); J. A. Appelbaum and W. F. Brinkman, Phys. Rev. 183, 553 (1969).

[3]W. A. Thompson, F. Holtzberg, and S. von Molnar, IBM J. Res. Develop. 14, 279 (1970).

[4]I. Giaever and H. R. Zeller, Phys. Rev. Lett. 20, 1504 (1968); C. B. Duke, G. G. Kleinman, A. M. Andrews, R. D. Burnham, N. Holonyak, Jr., and H. W.

Korb, in *Proceedings of the Tenth International Conference on the Physics of Semiconductors, Cambridge, Massachusetts, 1970,* edited by S. P. Keller, J. C. Hensel, and F. Stern, CONF-700801 (U. S. AEC Division of Technical Information, Springfield, Va., 1970), p. 856.

[5]J. G. Adler and T. T. Chen, Solid State Commun. 9, 501 (1971).

[6]J. M. Rowell, J. Appl. Phys. 40, 1211 (1969).

[7]W. L. McMillan and J. M. Rowell, in *Superconductivity*, edited by R. D. Parks (Marcel Dekker, New York, 1969), Chap. 11.

[8]J. A. Appelbaum and W. F. Brinkman, Phys. Rev. 186, 464 (1969).

[9]R. J. Birgeneau, J. Cordes, G. Dolling, and A. D. B. Woods, Phys. Rev. 136, 1359 (1964).

[10]R. E. Dietz, G. I. Parisot, and A. E. Meixner, Phys. Rev. B 4, 2302 (1971).

[11]M. T. Hutchings and E. I. Sammelsen, Solid State Commun. 9, 1011 (1971).

[12]R. E. Dietz, W. F. Brinkman, A. E. Meixner, and H. J. Guggenheim, Phys. Rev. Lett. 27, 814 (1971).

Destruction of Ferromagnetism in $ZrZn_2$ at High Pressure

T. F. Smith*
Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, California 92037

and

J. A. Mydosh
Institut für Festkörperforschung, Kernforschungsanlage, 517 Jülich 1, Germany

and

E. P. Wohlfarth
Department of Mathematics, Imperial College, London, England
(Received 12 October 1971)

The variation of the Curie temperature T_c for the weak ferromagnet $ZrZn_2$ has been determined as a function of hydrostatic pressure up to 25 kbar. T_c is found initially to decrease linearly with pressure at a rate $\partial T_c/\partial P = (-1.8 \pm 0.04) \times 10^{-3}\,{}^\circ\mathrm{K\ bar}^{-1}$, followed by an abrupt change of slope at 7.5 kbar and a rapid fall, extrapolating to a critical pressure P_c of 8.5 kbar for the destruction of ferromagnetism. The measured $T_c(P)$ curve agrees well with the relationship $T_c(P) = T_c(0)(1 - P/P_c)^{1/2}$ which is derived from the theory of itinerant ferromagnetism.

The Laves phase compound $ZrZn_2$ is considered to be a prime example of a weak itinerant electron ferromagnet.[1] This compound is composed of two elemental superconductors, neither of which possess the usual electronic configurations ($3d$, $4f$, or $5f$) found in magnetic materials. A small effective magnetic moment ($\sim 0.15\ \mu_B$ per Zr atom) and a low Curie temperature (~ 20°K) point towards the itinerant nature of this compound.[2] It is also expected for itinerant electron systems that large pressure effects will be observed in the magnetic properties.[3] Indeed, our measurements of the Curie temperature of $ZrZn_2$ indicate the destruction of the ferromagnetic state for pressures in excess of 8.5 kbar. This represents the first known example of a ferromagnetic system in which the magnetic behavior can be, apparently, completely suppressed by the application of pressure.

The sample of $ZrZn_2$ consisted of three pieces (total mass ~140 mg) broken from a dense conglomerate of large crystallites (typical dimensions ~0.1 cm) which had been grown[4] by the vapor transport technique. An x-ray examination[5] indicated that the material is single phase and has the C15, Laves phase structure with a lattice parameter of 7.396 ± 0.001 Å. The onset of the ferromagnetic state was detected by means of an ac mutual-inductance technique in which the off-balance signal from a pair of matched and opposed secondaries, one of which surrounds the sample, is continuously monitored as a function of temperature. The applied 150-Hz signal is estimated to produce a peak-to-peak field at the sample of ~0.2 G. The inductance measurements were made in a clamp device, in which pressure applied at room temperature using a hydrostatic pressure medium of a 1:1 mixture of n-pentane and isoamyl alcohol may be contained, permitting the removal of the high-pressure stage from the