competition from other effects, and alteration of the Υ well by the finite size of a contained plasma core. In regard to this last point, consideration of the analog experiment[7] (unpublished results) and the levitation experiment[13] indicates that an Υ well exists for test-sphere diameters up to about $r_0/R \leq 0.1$ in the present case.

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ASPHERICAL 3*d* ELECTRON DISTRIBUTION IN Ni^{++}†

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The purpose of this Letter is to report the results of the first measurement of an aspherical magnetic form factor of an antiferromagnetic single crystal, nickel oxide, using unpolarized neutrons. The simple structure, if this compound is purely ionic, makes it possible to explain the resultant e_g symmetry that is obtained. This differs from previous (polarized beam) measurements[1-4] where no simple explanation of the asymmetries is possible because the materials studied were either metals or (in one case) had the complicated spinel structure. The form factor of Ni^{++} is found to lie above the free-atom curve in contrast to the case of Mn^{++} where the converse is true. These results are of interest because of the very recent Hartree-Fock spin polarized calculations for Ni^{++} by Watson and Freeman.[5]

Unpolarized neutrons were used since the more accurate polarized neutron beam technique[6] is not applicable to antiferromagnets like NiO that do not have superimposed nuclear and magnetic reflections. The single crystal used for this investigation was cut in the shape of a cylinder (8 mm high, 2.3 mm in diameter) from a large boule grown by flame fusion. Integrated intensities of all nuclear and magnetic reflections were measured at room temperature up to a value of $\sin\theta/\lambda = 0.78$ using a wavelength of 1.046 A. From the heavily extinguished nuclear reflections a mosaic spread parameter of 112 seconds of arc was determined. This resulted in secondary extinction corrections to the magnetic intensities of only 12% for the strongest reflection (111), of only 3% for the next strongest ($\bar{3}11$), and of a completely negligible amount for all the other weaker reflections which occur at higher Bragg angles.

Effects due to double Bragg scattering became particularly noticeable when measuring the very weak outer reflections (signal to noise ratios in the range 0.05 to 0.1). In order to help correct for these effects, additional room temperature measurements were taken at a second wavelength of 1.005 A. By taking account of the changes that occurred in the peak shapes upon changing the

wavelength, it was possible to obtain estimates of the true intensity of the reflections under study. However, in many cases the error entailed in this procedure was rather large.

The experimental values for the magnetic form factor are plotted in Fig. 1 as the circles. The values were put on an absolute scale by using the previous result[7] that the value of the form factor for the (111) reflection is 0.94. The important observation to be made is that values of the form factor are obtained which depart radically from a smooth curve. In particular, outer reflections which occur at the same Bragg angles show very different values for their form factors. The existence of such a multivalued form factor demonstrates that the unpaired $3d$ electrons are distributed aspherically about each Ni^{++} ion.

To investigate the possibility (however unlikely) that the observed results might be due to the effect of anisotropic motions, either of the atoms with respect to one another or of the atoms with respect to their electron clouds, the $(\bar{1}\bar{1}, 1, 1)$ and $(\bar{5}, 7, 7)$ reflections, which occur at the same Bragg angle, were measured after cooling the crystal to 117°K. If the effect were due to anisotropic vibrations one would expect the ratio of the form factors of the two reflections to approach approximately unity as the temperature approaches 0°K. Within the limits of experimental error, this ratio did not change upon cooling from 297°K to 117°K.

The aspherical unpaired $3d$ electron distribution observed experimentally is just what is expected due to the splitting of the energy levels of the d electrons in the field of the anions. According to crystal field theory,[8] in an electric field of octahedral symmetry the five-fold degenerate levels split into a low-lying triplet of t_{2g} symmetry and a higher doublet of e_g symmetry. Since the levels fill up according to Hund's rule, the two unpaired electrons which give rise to the magnetic moment should then exhibit pure e_g symmetry.[9]

The form factor can be written as the sum of a spherical part and an aspherical part, $f = f_S + f_A$. The part f_A (having pure e_g symmetry in our case) can be calculated following the methods of Weiss and Freeman.[10] However, in order to compare the experimental results directly with those predicted for an f_A of e_g symmetry, it is necessary first to determine f_S. Since at the present time there is no adequate way of calculating f_S for atoms bound in a solid, f_S was chosen to yield the best agreement between the experimental and calculated values of the form factor. It was found to be possible (also most convenient and instructive) to do this by scaling the free-atom spherical form factor[11] for Ni^{++}, i.e., $f_S(x) = f_0(kx)$, where f_0 is the free-atom form factor, $x = \sin\theta/\lambda$, and the scale factor $k = 0.83$. In Fig. 1, f_S is plotted as the solid curve.[12] The final calculated values of f are denoted by the

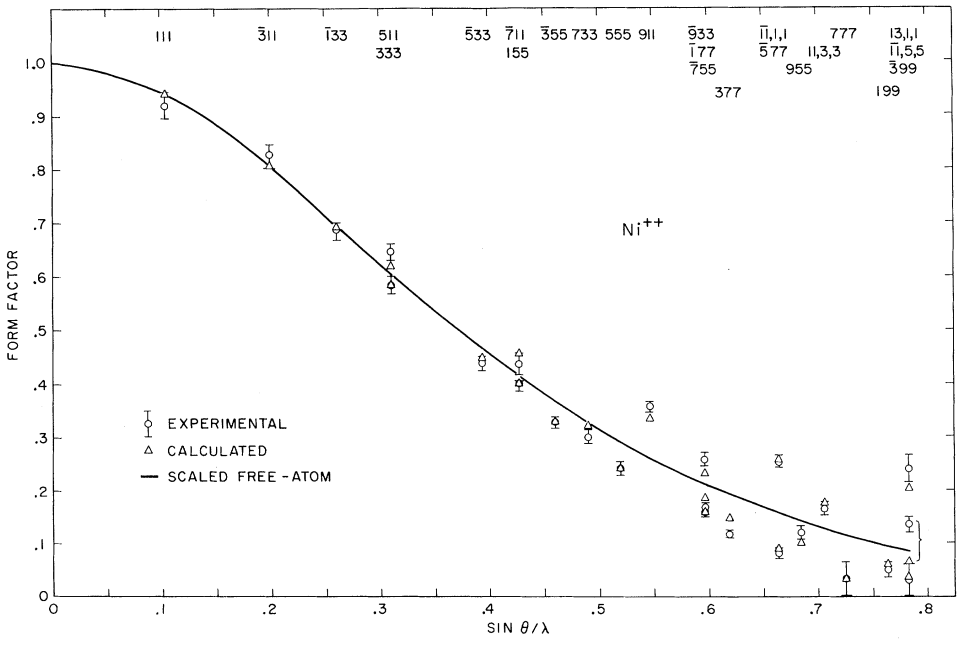

FIG. 1. The magnetic form factor of Ni^{++} in nickel oxide. The indices at the top of the figure refer (in the order shown) to the reflections directly below.

triangles. Reasonable agreement is thus obtained between the experimental form factor values and those computed assuming the unpaired electrons to be in a state of pure e_g symmetry.

It is important to observe that the scale factor, k, here determined experimentally, is such as to make the unpaired $3d$ charge distribution for Ni^{++} much more compact in the solid than it is for the free atom. This is in contrast to the case of Mn^{++} where experiments[13] show that the charge distribution is expanded in the solid.

These experimental results may be compared with the recent calculations of Watson and Freeman[5] for the Ni^{++} ion. These Hartree-Fock self-consistent field calculations allow the wave functions of electrons with opposite spins to have different radial dependencies (spin polarization) and lead to a contraction of the charge distribution (both for the free-atom case and the case where the Ni^{++} ion is placed in an octahedral array of point charges).[14] Unfortunately, the magnitude of the contraction is much too small to explain the observations reported here. Nevertheless, the important fact that the relation of the observed f_S to the free-atom f_S is just opposite for the cases of Ni^{++} and Mn^{++} would lead one to look for the origin of this effect in the outstanding difference between the two ions: namely their differing spin configurations. These experimental results also serve to suggest that whereas effects such as spin polarization and crystalline environment have a large influence on f_S, their effect on f_A is small.

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TUNNELLING FROM A MANY-PARTICLE POINT OF VIEW*

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Giaever[1] and more recently Nicol, Shapiro, and Smith[2] have observed the tunnelling current flowing between two metals separated by a thin oxide layer. The most interesting results are obtained when one or both of the metals are superconducting, in which case they find direct evidence for a gap in the quasi-particle spectrum of the superconductor. They were able to account for the data quantitatively on the assumption that the only relevant factor is the density of states in energy. This is to be expected if the transition probability for transfer of an electron from one side to the other is given by the familiar expression $(2\pi/\hbar)|M|^2\rho_f$, where M is the matrix element and ρ_f the energy density of final states, and if it is further assumed that M can be treated as a constant. It is implied that M is not only independent of energy for the small energy differences involved, but is also unchanged when the metal goes from normal to superconducting.