most to the conductivity. Moreover, the mobility edge has little connection to the optical absorption as suggested in Ref. 1.

[22] A model recently proposed by M. Cohen yielding a similar result (Ref. 2) poses difficulties since he assumes carrier percolation with a "mean free path" short compared to the distance between barriers, and that the barriers are produced by space-charge fluctuations, causing potential valleys of ~0.1 eV. These difficulties force the assumption (Ref. 2) of a long equilibrium distance between hole and electron quasi-Fermi levels of ≃1 μm, and deal with the fact that localization in shallow potential valleys (0.1 eV) cannot be achieved (uncertainty relation).

MAGNON SPECTRUM OF ALPHA OXYGEN*

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The far-infrared absorption in solid alpha oxygen at 27 cm^{-1} is shown to be definitively associated with the magnetic rather than orientational ordering in the crystal. A second magnetic excitation has been found at 6.5 cm^{-1}. The properties of both absorptions are described using the excitation-wave formalism.

It has been well established that structural phase transitions occurring at low temperatures in simple diatomic molecular solids are associated with the rotational degrees of freedom of the molecule.[1] In particular, the α-β transition in oxygen is coincident with the onset of precession of the molecules.[2] It is also coincident with the Néel point of the electronic spin system, clearly emphasizing the possibility of interdependence of the orientational and magnetic ordering in the crystal. With these considerations in mind, we have experimentally and theoretically re-examined the magnon spectrum of α oxygen. The excitation-wave method has been applied to a biaxially anisotropic magnetic structure to explain satisfactorily the energies, relative intensities, and magnetic-field dependence of magnons observed in our own and earlier work.

Molecular crystals such as α-N_2 are known to exhibit phonon absorptions in the far infrared with temperature-dependent frequencies and intensities near the transition from the α to the β phase.[3] Therefore, for α-O_2, observation of temperature-dependent behavior for the 27-cm^{-1} absorption[4] is not sufficient evidence to associate it with magnetic excitation. Absence of an isotopic mass shift would be sufficient. Thus a sample of 99.2% isotopically pure α-$^{18}O_2$, 3.1 mm thick, was grown, and its far-infrared spectrum measured at 1.6°K. The frequency shift of ~1.5 cm^{-1} expected for the 27.5-cm^{-1} line, had it been a phonon, was not observed.[5]

It should now be possible to construct a phenomenological spin Hamiltonian which would predict the 27.5-cm^{-1} magnon. On the basis of the monoclinic symmetry ($C_{2h}{}^3$) of the crystal lattice[6] and the assumption of isotropic exchange, we write

$$H=\sum_i (A S_{ix'}{}^2+B S_{iy'}{}^2)+\sum_{i>j} J\vec{S}_i\cdot\vec{S}_j .$$

J is the isotropic antiferromagnetic exchange constant, and $\pm z'$ is taken to be the direction of the moments. The subscript i indexes all molecules and the subscript j indexes the eight near neighbors of the ith molecule. A and B characterize the "single-molecule" anisotropy for which there are at least two sources. The magnetic dipole-dipole interaction between the two unpaired electronic spins in the free oxygen molecule produces an anisotropy energy of the form $AS_{x'}{}^2$.[7] $S_{x'}$ is the projection of the total molecular electronic-spin angular momentum ($S=1$) on the internuclear axis. Since A has been shown to be positive,[7] the spin S in the ground state is confined to a plane perpendicular to the internuclear axis. An assumption of this note is that the planar localization is not significantly disturbed when the molecule is placed in the α crystal lattice.[8] A second, in-plane, anisotropy is needed to stabilize the antiferromagnetic ordering of the α solid. Since the site symmetry of the oxygen molecule is C_{2h}, the twofold directional degeneracy of Π electronic states is removed. The nonzero off-diagonal elements of the total electronic orbital angular momentum between the Σ ground state and the excited Π states will reflect this anisotropy, which in turn will affect the spin orientation via the spin-orbit coupling. Because of the smallness of the spin-orbit coupling constant,[7] such a term is expected to be small, but nevertheless, important.

According to this picture, the sublattice magnetization vectors lie in the crystallographic a-b plane which is normal to the molecular axes.[6] The precise orientation has yet to be determined, but neutron-diffraction data do not preclude this general arrangement.[9] Following the basic outline by Trammell,[10] diagonalization of the Hamiltonian produces the following expressions for the magnon energies:

$$E^+(k)^2=(\alpha+\beta)^2-\gamma^2,\quad E^-(k)^2=(\alpha-\beta)^2-\gamma^2,$$

where

$$\alpha=\frac{A+B}{2}+\left[\frac{(A-B)^2}{4}+(8J\langle S_{z'}\rangle)^2\right]^{1/2},$$

$$\beta=4J\sin2\theta\left[\cos\left(\frac{k_xa}{2}\right)\cos\left(\frac{k_yb}{2}\right)+\cos\left(\frac{k_yb}{2}\right)\cos\left(k_cC-\frac{k_xa}{2}\right)\right],$$

$$\gamma=4J\left[\cos\left(\frac{k_xa}{2}\right)\cos\left(\frac{k_yb}{2}\right)+\cos\left(\frac{k_yb}{2}\right)\cos\left(k_cC-\frac{k_xa}{2}\right)\right]$$

The magnitude of θ is a measure of the mixing of the $m_s=\pm1$ states by anisotropy. a, b, c are lattice parameters. The theory predicts the existence of two magnon branches. The ratio of absorption intensities for the $k=0$ modes is

$$\frac{I^-}{I^+}=\frac{E^-}{E^+}\,\frac{\{\alpha+\beta+\gamma+[(\alpha+\beta)^2-\gamma^2]^{1/2}\}\{\alpha-\beta-\gamma+[(\alpha-\beta)^2-\gamma^2]\}^{1/2}(1-\sin2\theta)}{\{\alpha+\beta-\gamma+[(\alpha+\beta)^2-\gamma^2]^{1/2}\}\{\alpha-\beta+\gamma+[(\alpha-\beta)^2-\gamma^2]\}^{1/2}(1+\sin2\theta)}.$$

For reasonable values of the parameters, the absorption intensity of the higher frequency line is greater than that of the lower.

The line at 27 cm^{-1} is the most intense absorption in α oxygen in the far infrared. It is easily observed in samples the order of 1 cm thick. In light of the above considerations, a second absorption was sought at energies down to 5 cm^{-1}. Crystals of thicknesses varying between 4.4 and 8 cm were grown by slow cooling from the liquid to the α solid. Spectra were measured using a Michelson interferometric spectrometer and associated cryogenic apparatus described elsewhere.[11] Results for a 5-cm sample are shown in Fig. 1. Upon raising the temperature of the sample above the Néel point (23.8°K) the sharp absorption at 6.4 cm^{-1} disappeared. To associate this new absorption line with the antiferromagnetic order, data were also taken as a function of magnetic field. The polycrystalline samples were grown inside the bore of an 80-kG superconducting magnet. The results for the 6.4-cm^{-1} line as well as for the 27.5-cm^{-1} line are shown in Fig. 2.

On the basis of this experimental evidence, we assign 27.5 cm^{-1} to $E^+(0)$ and 6.4 cm^{-1} to $E^-(0)$. The measured integrated absorption intensity ratio $I^-/I^+=0.07\pm0.01$. Also known is the magnon energy at the edge of the Brillouin zone, obtained from sidebands in the optical double transition spectra of Litvinenko, Eremenko, and Garber.[12] The estimated value for E(zone edge) is 37.5 cm^{-1}.

The theory reproduces these energies and gives $I^-/I^+=0.05$ for $A=10.1$ cm^{-1}, $B=0.5$ cm^{-1}, and $J=4.0$ cm^{-1}. Dispersion curves for two high-symmetry directions[13] are shown in Fig. 3. The two branches become degenerate at the zone edge with $E=37.5$ cm^{-1} in both cases, as in fact they do at all high-symmetry points of the zone boundary. This explains the predominance of a single energy value in the data of Ref. 12. The magnitudes of A and B indicate

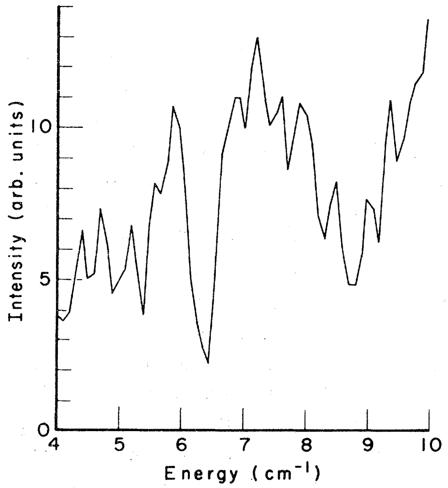

FIG. 1. Far-infrared absorption spectrum of a 5-cm sample of solid α oxygen. The envelope of the spectrum is a property of the energy source. The feature at 8.7 cm^{-1} is an interference fringe due to a filter in the light path. The resolution in frequency is 0.25 cm^{-1}.

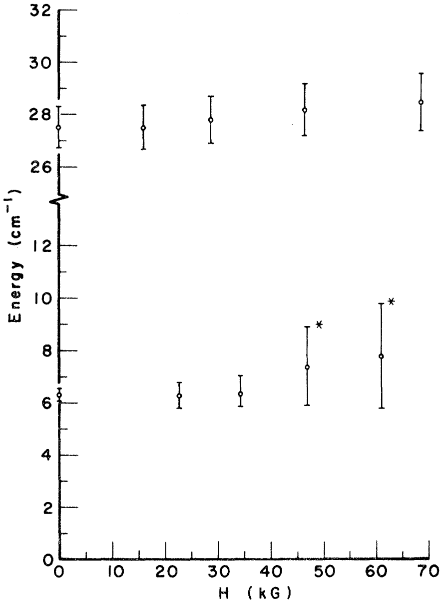

FIG. 2. Magnetic-field dependence of the two $k=0$ magnons. The circles mark the positions of maximum absorption and the bars indicate full width at half-absorption for spectra with frequency resolution of 0.7 cm^{-1}. The asterisks denote larger uncertainty in measurement than for the other data, due to the poorly defined line shape at the highest fields.

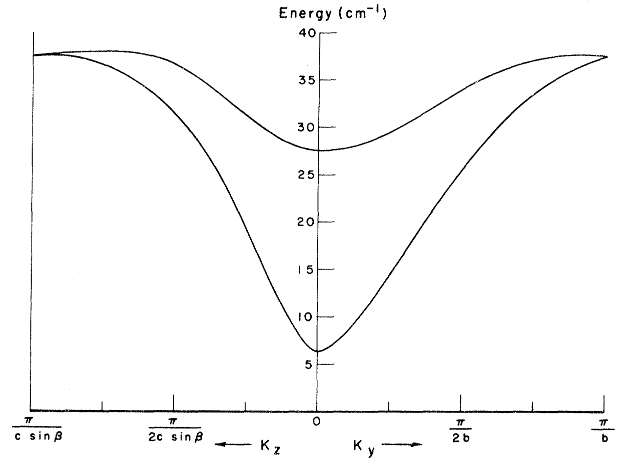

FIG. 3. Dispersion curves calculated from the excitation-wave theory for $A=10.1$, $B=0.5$, $J=4.0$. y is parallel to the twofold axis, which lies in the face centered plane, and z is the direction perpendicular to the face centered plane. (See Ref. 6.)

that in the crystal, mechanisms in addition to the ones we have discussed, such as anisotropic exchange and compression of the electron cloud, contribute to the effective "single-molecule" anisotropy. However, the system is still exchange dominated. $\langle S_{z'}\rangle$ is calculated to be 0.99, so the two sublattices are only slightly less than fully aligned. T_n obtained from the mean-field theory of Moriya[14] equals 31°K as compared with T_n(expt), 23.8°K. This discrepancy could indicate that indeed the long-range magnetic order would persist to higher temperatures than it does were it not disrupted by the loss of orientational order of the molecules at the α-β phase transition. This possibility has been indicated by Barrett and Meyer in their analysis of the crystal structures of oxygen.[15] Including a dc magnetic field in the excitation-wave Hamiltonian and assuming random distribution of crystallites reproduces qualitatively the observed nonlinear dependence of magnon energy and linewidth on applied field.

The two additional lines (44 and 37.5 cm^{-1}) observed by Blocker, Kinch, and West[4] and in the present series of experiments appear to be defect induced. The 44-cm^{-1} line corresponds to one of the two librational modes observed in the Raman spectrum of α oxygen.[16] If the crystal were perfect, this mode would be infrared inactive. The intensity of the other line did not scale with the thickness of the sample, was unshifted in the isotope experiment, and was significantly broadened by the magnetic field. Therefore we attribute it to defects at the sample surface, inducing magnon absorption at the zone edge.

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LARGE-ANGLE p-d SCATTERING AT 580 MeV

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The elastic scattering of protons from deuterons has been measured at 580 MeV. The results for large-angle scattering are presented and discussed here. The backward peak observed in the experiment is consistent with a baryon-exchange mechanism including the transfer of baryon resonances.

A pronounced increase in the cross section at large angles has been observed for proton-deuteron scattering in the energy range 1-1.5 GeV.[1,2] This back peak is also seen in the present experiment.[3] The magnitude of the peak is found to decrease rapidly with increasing proton energy.

The explanation of this effect most likely involves the transfer of a baryon, that is a pickup mechanism, because the momentum transfer required at the deuteron vertex is much smaller than that for a single or double impulse collision. This is also seen in a number of calculations. At 1 GeV the impulse series for back-scattering gives results more than an order of magnitude smaller than experiment,[4] while a dispersion relation calculation which includes the transfer process obtains a peak of the correct order of magnitude.[5] Using the $pp \rightarrow \pi d$ experimental results as the input, Craigie and Wilkin[6] have recently treated the back-scattering with a model that includes a baryon-transfer mechanism and also obtained the correct order of magnitude for the back peak at 1-1.5 GeV.

Kerman and Kisslinger[7] have studied the large-angle p-d cross section for 1-GeV protons as a baryon-transfer reaction. They find that the S-state component of the deuteron wave function, which by itself is satisfactory for the calculation of nucleon-transfer reactions at low energies, gives a negligibly small contribution at 1 GeV. Using a variety of wave functions with 6-7% D state, only about 50% of the cross section can be