resonances in the normal magnetoacoustic effect arise from these matrix elements, which in the semiclassical limit reduce to Bessel functions[2] of order $(n'-n)$ whose argument is $(q_y v_0/\omega_0) \times (n/n_0)^{1/2}$. The parallel field magnetoacoustic resonances discussed in this work, like the normal magnetoacoustic resonances, are *semiclassical* in nature. The reduction of Eqs. (7)-(11) to the semiclassical limit[2] will again give Bessel functions in the conductivity tensor.

From the similarity in the conductivity tensor, we may assume that the resonances in the parallel field case arise in the same way as in the normal magnetoacoustic effect. The resulting condition for a maximum in attenuation is

$$2[(2\hbar c n_0/|e|B_0)\alpha_4^2/(\alpha_1\alpha_2^3)^{1/2}]^{1/2}=l\lambda, \qquad (12)$$

where l is any positive integer, λ the wavelength of the sound wave, and n_0 the quantum number of the last occupied Landau level.

It is quite simple to demonstrate that the quantity on the left-hand side of Eq. (12) is equal to the "oscillation distance," $\frac{1}{2}\oint|v_z|dt$, for the electrons on orbit A of Fig. 1. As we remarked previously, these electrons oscillate back and forth parallel to $\vec{B}_0$ as they go through one full cyclotron orbit. The condition for minimum attenuation would be that the "oscillation" distance equal a half-integral number of wavelengths. We should like to point out that these resonance conditions are quite different from those assumed by Daniel and Mackinnon,[6,7] although the calculations confirm the intuitive guess that anisotropy of the Fermi surface gives rise to this effect.

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[1] L. Mackinnon, M. T. Taylor, and M. R. Daniel, Phil. Mag. 7, 523 (1962).

[2] J. J. Quinn and S. Rodriguez, Phys. Rev. 128, 2487, 2494 (1962).

[3] M. H. Cohen, M. Harrison, and W. A. Harrison, Phys. Rev. 117, 937 (1960).

[4] V. L. Gurevich, V. G. Skobov, and Yu. A. Firsov, Zh. Eksperim. i Teor. Fiz. 40, 786 (1961) [translation: Soviet Phys.–JETP 13, 552 (1961)].

[5] J. J. Quinn (to be published).

[6] M. R. Daniel and L. Mackinnon, Phil. Mag. 8, 537 (1963). In this paper the authors assume that

$$\oint v_z dt=l\lambda$$

is the condition for maximum absorption. They show that

$$\oint v_z dt=-(c\hbar/|e|B_0)(\partial A/\partial k_z),$$

where A is the cross-sectional area of the orbit in k space. The electrons which we believe to be responsible for the resonances have $\partial A/\partial k_z=0$.

[7] E. A. Kaner, V. G. Peschanskii, and I. S. Privorotskii {Zh. Eksperim. i Teor. Fiz. 40, 214 (1961) [translation: Soviet Phys.–JETP 13, 147 (1961)]} have made some consideration of the propagation of sound in anisotropic crystals. For the case of propagation parallel to the magnetic field, it seems that their resonance condition would be similar to that of reference 6 instead of the condition of the present paper.

OPTICAL MASER OSCILLATION FROM Ni^{2+} IN MgF_2 INVOLVING SIMULTANEOUS EMISSION OF PHONONS

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(Received 29 August 1963)

All of the solid state optical maser materials known at the present time take advantage of purely electronic transitions in solids. We wish to report the observation of fluorescence from Ni^{2+} in MgF_2 and optical maser oscillation in a line which involves a transition between electronic states of Ni^{2+} and simultaneous vibrational excitations of the MgF_2 lattice.

The polarization of infrared emission from Ni^{2+} in MgF_2 at 20°K is shown in Fig. 1. The sharp structure near 6500 cm^{-1} represents pure electronic transitions between the first excited state $^3T_2$ and the ground state $^3A_2$,[1] while the remainder of the spectrum arises from electronic transitions with simultaneous emission of phonons to the lattice. As can be seen from Fig. 1, the π spectrum coincides with the axial spectrum and both the electronic and phonon-accompanied transitions are, therefore, magnetic dipole. The magnetic-dipole character of the purely electronic transitions in MgF_2:Ni^{2+} has been reported earlier.[2] The intense narrow emission at 6500 cm^{-1} consists of three closely spaced lines representing transitions from the lowest $^3T_2$ level to the three components of the $^3A_2$ ground state at 0, 1, and 6 cm^{-1}. The 1-cm^{-1} separation is barely re-

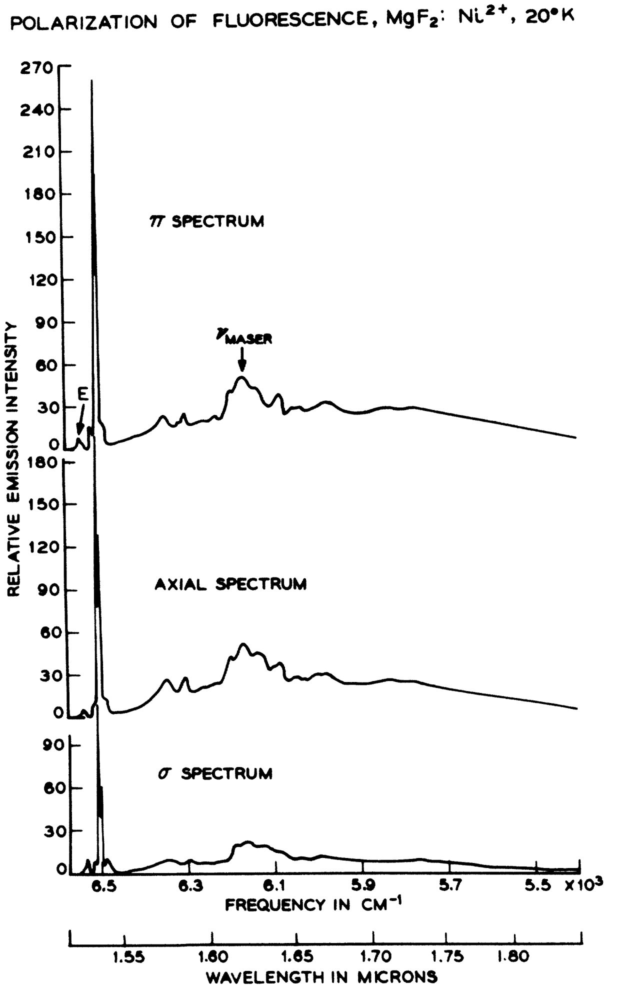

FIG. 1. The polarization of fluorescence from Ni^{2+} in MgF_2 at 20°K. The sharp structure above 6500 cm^{-1} arises from pure electronic transitions. Maser oscillation occurs at the frequency of maximum emission on the vibrational part of the spectrum.

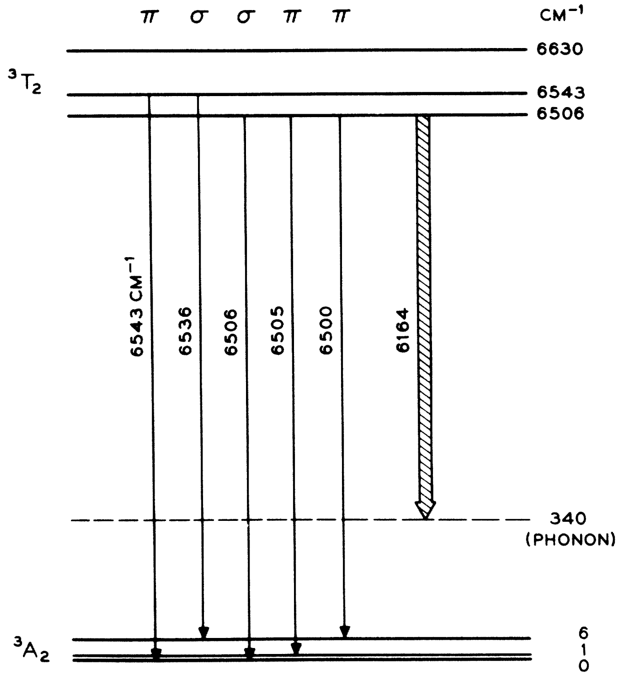

FIG. 2. The energy levels of Ni^{2+} in MgF_2 assigned from the fluorescence spectra of Fig. 1. The three components of the $^3A_2$ ground state and three of the nine components of the $^3T_2$ first excited state are identified. The maser frequency and the phonon energy are also included but these are associated with the crystal as a whole (see text).

solved at 20°K. More precise data on Ni^{2+} in ZnF_2 places the ground-state components at 0, 1.5, and 6.8 cm^{-1}.[3] At slightly higher energy, two components of a weak electronic transition to the ground state are resolved at 20°K (labeled E in Fig. 1). These originate from a $^3T_2$ level 37 cm^{-1} above the initial state for the strong triplet. The polarization character of these transitions and the energy levels involved are shown in Fig. 2. Due to the weakness of the line, we are unable to determine from the spectra which of the lowest two components of $^3A_2$ is the terminal state for the π line at 6543 cm^{-1}. Assignment to the lowest component is made on the basis of selection-rule calculations for magnetic-dipole transitions in this system.[4] An emitting state at about 6630 cm^{-1} is seen weakly in π polarization at 77°K. Thus three of the nine components of the $^3T_2$ state of Ni^{2+} in MgF_2 are seen in fluorescence. At room temperature, emission consists of a broad band centered at 6100 cm^{-1} extending from 5000 to 7500 cm^{-1}. Prominences due to the electronic lines are absent. The measured fluorescence lifetime at 77°K is about 10 msec.

The unique feature of stimulated emission from Ni^{2+} in MgF_2 is that maser oscillation is observed in the phonon wing of the emission spectrum and not in the sharp electronic lines (see Fig. 1). Stimulated emission is characterized by the familiar "spiking" phenomenon. The spectral distribution of maser emission was obtained by directing the maser beam onto the entrance slit of a grating spectrometer and measuring the integrated power output under pulse illumination from an FT524 xenon lamp within a spectral range of 0.5 cm^{-1} about a fixed wavelength. The measurement was then repeated successively at slightly different wavelengths to obtain a point-by-point mapping of the spectral profile. At 20°K maser emission appears in a line centered at 6164 cm^{-1} with a width of about 6 cm^{-1}. At 77°K the line shifts to lower energy by about 4 cm^{-1}, the measured width is 4 cm^{-1} (less than at 20°K), and the threshold is 12% higher.

Although emission is observed initiating from two of the $^3T_2$ states at 77 and 20°K, we associate the phonon energy identified by the maser experiment with electronic transitions from the lowest $^3T_2$ component, both because the intensity of these transitions is stronger and because the threshold decreases with decreasing temperature. The phonon energy thus derived is about 340 cm^{-1} (Fig. 2). The shift of the maser line with temperature is not inconsistent with the shift of the electronic lines, although a precise comparison is difficult due to line broadening at 77°K. The narrower width of the maser line at the higher temperature (77°K) might appear to be inconsistent. However, although the crystals are immersed, crystal temperature is undoubtedly higher than the bath temperature during the light pulse, and for the same level of input power the temperature rise would be greater at 20°K due to a reduced heat capacity and lower thermal conductivity. The widths reported here, therefore, do not represent the limit of line narrowing but must be considered upper bounds imposed by the power-input condition required to achieve maser oscillation.

The phonon energy and the maser frequency are included on the energy-level diagram of Fig. 2, but these should not be interpreted as an energy level and a transition belonging to the Ni^{2+} ion. The maser frequency results from a lattice excitation plus an electronic transition to the ground state of the ion, i.e., a transition of the crystal as a whole. Therefore, although the ion makes a transition to the ground state, the terminal state for the maser is an excited state of the lattice-impurity system. It might be argued that oscillation in the strong and sharp purely electronic lines should be more favorable. However, absorption losses in these resonance lines are high while absorption at the maser frequency is negligible. Since oscillation is not observed in the electronic lines, it follows that maser action occurs in the region of phonon emission before the ion population is inverted.

Two different proposals may be suggested concerning the frequency of maser oscillation on the vibrational continuum. The first is that the oscillation frequency is determined by a selective preference for the emission of a 340-cm^{-1} phonon. However, it is difficult to find valid reasons why such a preference should occur. The second proposal (and that which underlies our preceding remarks) is that oscillation occurs at the maximum of the gain versus frequency curve for the system.[5] Since absorption losses are negligible over most of the range spanned by the phonon part of the spectrum, maximum gain occurs at the frequency where ν^{-4} times the spontaneous emission intensity is a maximum (the ν^{-4} factor gives the density-of-states and spectrometer corrections necessary to convert from the recorded spontaneous emission to stimulated emission strength). The assignment of a particular phonon to this maximum is justified only because the maximum in the emission spectrum is a well-defined peak which coincides with the maximum of the gain curve. However, the association of a phonon with the maser transition in no way implies that this is the only phonon assisting in maser oscillation. The fluorescence spectrum clearly shows that many overlapping vibrational modes of the crystal are excited. Therefore, all modes contributing to the intensity maximum are involved.

It has been pointed out to the authors by D. E. McCumber, who will publish a detailed analysis of the role of phonon coupling in maser systems of the type reported here, that a choice between these two alternatives may be possible with the aid of wavelength-selective reflectors which, in the gain-curve model, would be capable of continuously shifting the frequency of maser oscillation over a broad, smooth emission curve.

We wish to express our deep appreciation to D. E. McCumber and C. G. B. Garrett for many stimulating discussions which contributed significantly to the ideas expressed in this paper. We also wish to thank R. A. Thomas for assistance with the experimental work.

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[1] See, for example, K. Knox, R. G. Shulman, and S. Sugano, Phys. Rev. 130, 512 (1963).

[2] J. Ferguson, H. J. Guggenheim, L. F. Johnson, and H. Kamimura, J. Chem. Phys. 38, 2579 (1963).

[3] M. Peter and J. B. Mock, Phys. Rev. 118, 137 (1960).

[4] H. Kamimura (private communication).

[5] D. E. McCumber, Phys. Rev. 130, 675 (1963).