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[10]A Lorentz-broadened vibrator characterized by a known frequency and width, by mass m_H, and by charge $|q|=e$ would have σ_{peak} much larger and T_1 so small as to be quite inconsistent with the observed width. The strength-integral value cited corresponds to $|q|=0.18e$. Note that $|q|$ enters as the square.

Magnetization Induced by Optical Pumping in Antiferromagnetic MnF_2†

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We have generated magnetization in antiferromagnetic MnF_2 by selectively pumping the lowest $^6A_1 \rightarrow {}^4T_1$ sublattice excitons (or exciton-magnon band) with a pulsed, tunable dye laser. Intersublattice relaxation processes were studied by observing the creation and decay of specific excitations, as induction signals. The exciton relaxation time decreases with increasing temperature and pumping power. We interpret this as intersublattice relaxation assisted by thermally excited magnons.

We have generated magnetization in antiferromagnetic MnF_2 by selectively creating sublattice excitons and magnons with a pulsed, tunable dye laser. The magnetization changes, and those due to subsequent relaxation processes, were observed as induction signals. This enables us to measure, in the time domain, very small intersublattice couplings (~10^{-5} cm^{-1}), of the type leading to Davydov splitting. The technique promises to be useful for studying the dynamics of specific excitations which are resonantly created by pumping with tunable lasers.

The magnetic moment of a two-sublattice antiferromagnet which is excited to one of its exciton (or magnon) levels $\Gamma_{A,B}$ is

$$M(t)=[n_A(t)-n_B(t)]\langle\Gamma_A|\vec{\mu}|\Gamma_A\rangle, \tag{1}$$

where $n_A(t)$ and $n_B(t)$ are the numbers of excitons on sublattices A and B, respectively, at time t, and $\vec{\mu}$ is the magnetic dipole operator. The possibility of having $n_A(t)\neq n_B(t)$ depends on two main factors: (i) A scheme (in this case optical pumping) can be devised to populate one sublattice preferentially. This requires that any magnetic domains have substantially the same orientation. (ii) The transfer of excitation (TOE) between sublattices must be slower than, or comparable to, the observation time. Experiments on magnetization induced by optical pumping have been reported for paramagnetic systems.[1,2] The physical mechanisms operating in these cases are quite different from those responsible for what we observe in antiferromagnetic MnF_2.

In the present work we resonantly excite the lowest magnetic dipole $(\Gamma_1{}^+, \Gamma_2{}^+)$ exciton ($E1$) in MnF_2 at 18 418 cm^{-1} or the associated electric dipole exciton-magnon band at 18 476 cm^{-1}. These arise from the $^6A_1 \rightarrow {}^4T_1$ transition of the Mn^{2+} ions.[3] Selective pumping of the excitons is made possible by applying [110] stress which lifts the sublattice degeneracy. The laser is then tuned to excite a particular sublattice exciton[4] Γ_A or Γ_B, where

$$\sqrt{2}|\Gamma_A\rangle=|\Gamma_1{}^+\rangle-|\Gamma_2{}^+\rangle \quad \text{and} \quad \sqrt{2}|\Gamma_B\rangle=|\Gamma_1{}^+\rangle+|\Gamma_2{}^+\rangle. \tag{2}$$

Stress measurements by Dietz[5] showed that any Davydov splitting of $(\Gamma_1{}^+, \Gamma_2{}^+)$ is much less than the linewidth (0.5 cm^{-1}). Measurements of the thermalization of the intrinsic emission from $E1$ show that when the sublattices are out of resonance by 5 cm^{-1} the TOE rate between sublattices is less than 10^4 sec^{-1} below 2°K.[6] Thus for a single-domain crystal the conditions (i) and (ii) can be satisfied.

The situation is somewhat more complicated in the case of exciton-magnon pumping. Under [110] stress the exciton-magnon band splits into components excited by $\vec{E}\parallel[110]$ and $\vec{E}\parallel[1\bar{1}0]$, corresponding to an exciton on sublattice A and a magnon on sublattice B and *vice versa*. This selection rule is a consequence of the short range of the interion exchange coupling[3,7] so that the interaction has higher symmetry than that of the crystal. If the exciton and magnon interchange sublattices slowly enough, single-sublattice excitons (together with the opposite sublattice magnons) can be created by linearly polarized light. The magnetic moment of the exciton-magnon state is very small ($\approx 0.02\mu_B$/ion)[8] so that almost no magnetization is created during its excitation. Then, for example, if the magnon relaxes separately from the exciton in a time much less than the duration of the laser pulse, a magnetization will be produced corresponding to exciton creation. We will see later that this is what happens.

The experimental arrangement is as follows: A pulsed, tunable dye laser is focused to ~0.05 cm diam in a crystal of MnF_2 $0.2\times 0.2\times 1$ cm^3, held in a Dewar of pumped He^4 at 2°K. A pickup coil around the crystal detects the rate of change of magnetization. The laser is flash-lamp pumped and uses Na-fluorescein as the active medium. Its output is about 7 mJ/pulse and the total pulse duration is 0.6 μsec. Details of the laser construction have been published elsewhere.[9] The power density in the crystal is such that about 0.005% of the Mn^{2+} ions within the volume of the focused light are excited when $E1$ is pumped. Zeeman measurements[3] on $E1$ show that $|\langle\Gamma_A|\mu_z|\Gamma_A\rangle| = 2.11\mu_B$/ion and it follows from group theory that $\langle\Gamma_A|\mu_{x,y}|\Gamma_A\rangle = 0$. For $n_B = 0$, a magnetization of ~0.1 G is generated in the [001] direction. The emf induced in the pickup coil for a rate of excitation of 10^6 sec^{-1} is then on the order of 0.2 mV. Two pumping configurations were used. In the first, which we call the transverse case, the laser propagates along [110] with $\vec{H}\|[001]$ and $\vec{E}\|[1\bar{1}0]$. This is used to pump the σ-polarized excitons and one polarization of the exciton-magnon band. The moment is generated perpendicular to the long direction in this crystal, so coupling of the changing moment to the 50-turn pickup coil is enhanced by using a flux path of laminated Supermalloy. Stress was applied parallel to the laser direction [110] by a pneumatic piston acting through fused-quartz pads. The second, or longitudinal, configuration uses a crystal cut from the same part of the boule with its long dimension parallel to [001]. In this case the $\vec{E}$ vector of the light is along either [110] or $[1\bar{1}0]$, and the moment couples efficiently to a 100-turn coil whose axis is along [001]. The coils are connected directly to a field-effect transistor source-follower preamplifier in the He bath. Low-impedance cable transmits the signal to an external amplifier which is followed by an oscilloscope display. Data were taken on a single-shot basis. The background noise level was ~20 μV, and this limits the resolution of our present experiments. The emf induced in the pickup coil is

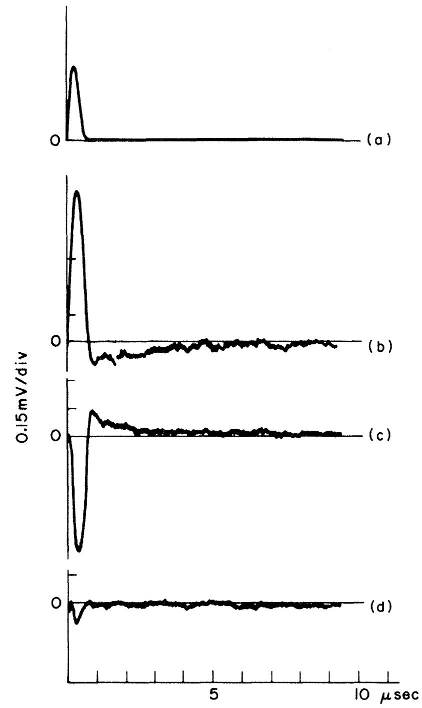

FIG. 1. Magnetic induction signals in MnF_2 associated with optical pumping of $^4T_1$ (Γ_1^+, Γ_2^+) excitons under [110] stress in the transverse configuration. (a) The dye laser pulse. (b) Pumping B-sublattice excitons. (c) Pumping A-sublattice excitons. (d) Out of resonance with the excitons.

$$e(t) \propto \frac{d}{dt}[M(t)] \propto W(t)G(0) - \int_0^t W(t')\frac{d}{dt}G(t-t')dt', \qquad (3)$$

where $W(t)$ is the rate of excitation and $G(t)$ is the response of the crystal magnetic moment to excitation by the laser. To obtain the actual signal observed, $e(t)$ should be convoluted with the response function of the coil and detection electronics. However, this was rarely necessary as the detection system had a response time about four times faster than the signals of interest.

The results obtained for exciton pumping are shown in Fig. 1. When Γ_A is excited, $n_A(t)\neq 0$ and $n_B(t)=0$, and a voltage pulse approximately in phase with the laser pulse and corresponding to exciton creation is observed. The absolute magnitude of this voltage was calibrated against a known moment change produced in a small test

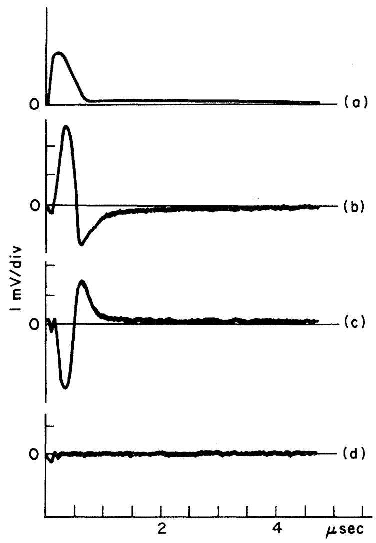

FIG. 2. Magnetic induction signals associated with optically pumping the exciton-magnon band in the absence of stress in the axial configuration. (a) The dye laser pulse. (b) Pump light polarized [110] to create excitons on sublattice B and magnons on A. (c) Pump light polarized [1$\bar{1}$0] to create excitons on sublattice A and magnons on B. (d) Pump light polarized [100] which is still in resonance but does not selectively populate the sublattice excitations.

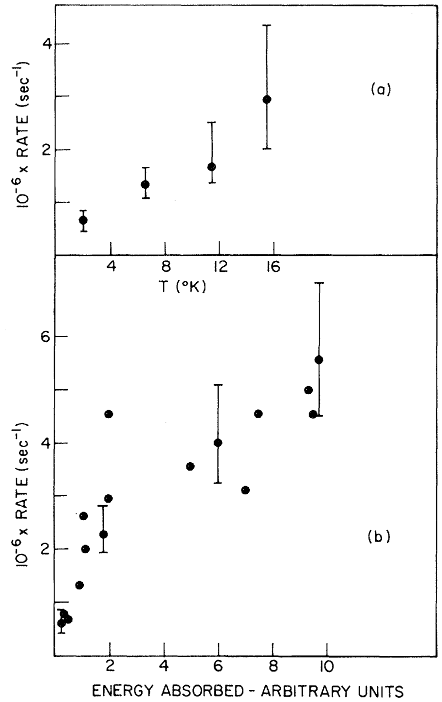

FIG. 3. (a) Temperature dependence of the magnetization decay rate τ_{s1}^{-1} assigned to exciton intersublattice relaxation. Data were taken with the laser power attenuated by a factor of 20 so that 0.1 mJ/pulse was absorbed in the exciton-magnon band. (b) Dependence of τ_{s1}^{-1} on the power absorbed in the crystal. Sample was immersed in liquid He^4 at 2°K.

solenoid. This agreed with the predictions of Eqs. (1) and (3) to better than a factor of 2, confirming the origin of the signal and showing that any crystal domains are substantially in the same direction. A voltage reversal corresponding to relaxation of the sublattice exciton moment is also observed (Fig. 1), with a relaxation time which decreases with increasing power. The mechanism for this relaxation will be discussed below. When the laser frequency is shifted slightly to become resonant with Γ_B, the sign of the induced voltages changes as expected from Eq. (1) since now $n_A(t)=0$ and $n_B(t)\neq 0$. No induction signals are observed when the laser frequency is moved off resonance with the exciton. This fact, together with the time duration of the voltage pulses and the polarization of the pump light, discriminates experimentally against the inverse Faraday effect.

The results obtained from pumping near the peak of the exciton-magnon band are shown in Fig. 2. As we have already noted, [110]-polarized pump light is expected to create excitons on sublattice A and magnons on B. The creation of this state is associated with a negligible change in the magnetization ($0.02\mu_B$/ion). The fact that a large induction signal is observed, essentially in phase with the pumping light, shows that the magnon created is either crossing sublattices or decaying, in a time less than ~50 nsec. This then simulates exciton creation and subsequent decay, so the results shown in Fig. 2 are similar to those in Fig. 1. (The reverse pulse again has a decay time which decreases with increasing pump power.) The sign of the induced voltage changes when the polarization of the pump light changes from [110] to [1$\bar{1}$0], since the excitations are being created on opposite sublattices. For [100]-polarized light, selective sublattice pumping is not achieved and no signals are observed. We have confirmed that the initial pulse in Fig. 2 has the sign and magnitude associated with exciton creation. This was done in the transverse geometry by pumping Γ_B directly, and then using [1$\bar{1}$0] polarization to pump the exciton-magnon exciting an exciton on B and a magnon on A.[5] We do not distinguish here between the possible magnon relaxation modes giving rise to the initial pulse.

Figure 3 shows the temperature and pumping-power dependence of the magnetization decay signals of Fig. 2. We assign this decay to intersublattice exciton relaxation with a rate τ_{s1}^{-1}. At the highest power or temperature, τ_{s1}^{-1} is ~4×10^6 sec^{-1}, compared to the steady-state value of ~10^4 sec^{-1} at 1.5°K.[6] The mechanism we propose for this temperature- and power-dependent relaxation is a spin-allowed scattering process in which an exciton and a thermally excited magnon[10] interchange sublattices. There is already evidence from the initial pulse in Fig. 2 that magnons created near the zone boundary equilibrate quickly between sublattices, and this will certainly be true of thermally excited magnons of general $\vec{k}$ since sublattice magnons are not eigenmodes.[11] Therefore, if excitons are created on one sublattice this scattering mechanism will lead to a decay of the exciton magnetization. The cross section for exciton-magnon scattering depends on both the number of magnons and the number of excitons excited. The magnon number ($\bar{n}_m$) increases with temperature[10] and causes τ_{s1}^{-1} to become larger [Fig. 3(a)], although the measured values of τ_{s1}^{-1} do not increase as rapidly as $\bar{n}_m(T)$. The exciton number increases with pumping power and this also leads to an increase in τ_{s1}^{-1} [Fig. 3(b)].[12]

It is worthwhile to consider briefly the reason for eliminating several other possible mechanisms. The number of photons emitted by trapped excitons was measured and found to be proportional to the pumping power. This eliminates stimulated photon processes as a cause of the exciton-decay signals. Exciton-exciton annihilation that produces a real intermediate state, which subsequently relaxes to a single exciton on the opposite sublattice, is eliminated for the same reason. Exciton-exciton scattering in which TOE between sublattices occurs in a virtual intermediate state is unlikely as it involves a $\Delta M_S = 2$ transition (since the excitons are created on the same sublattice), and it would not increase with increasing temperature. Other experiments performed on several different samples showed that impurity-induced exciton trapping was not responsible.

We have demonstrated the feasibility of generating detectable magnetization in an antiferromagnet by optical pumping, and in so doing confirm the extent of the sublattice nature of the magnons and $^4T_1$ excitons in MnF_2. We also show that information on relaxation processes of specific excitations can be obtained by this technique using modest pumping power. In particular the rate of exciton-magnon sublattice interchange has been determined as a function of temperature, and a lower limit of ~10^7 sec^{-1} placed on the magnon intersublattice relaxation.

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