Observation of antiferromagnetic resonance in epitaxial films of MnF_2

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Antiferromagnetic resonance has been observed in thin films for what we believe to be the first time. Epitaxial films of MnF_2 on MgF_2, ZnF_2, and FeF_2 substrates were used. The resonance positions were very close to, but slightly below, that extrapolated for the bottom of the spin-wave band in bulk MnF_2. The linewidths ranged from 15 to 500 Oe, with the narrowest ones corresponding to films with the best degree of epitaxy as judged by texture camera photographs.

INTRODUCTION

The fabrication of new materials by epitaxy has developed rapidly in recent years with the primary attention given to semiconducting[1] and metallic[2-4] superlattices. Relatively less effort appears to have been devoted to insulators—particularly magnetic ones. One notable exception has been the growth of yttrium iron garnet (YIG) films of high quality by liquid-phase epitaxy on garnets.[5-7] In both these and ferromagnetic metallic thin films and superlattices, ferromagnetic resonance (FMR) has been extensively studied.[8-10]

To our knowledge, no one has attempted to prepare thin films and/or superlattices of antiferromagnetic insulators such as MnF_2 and FeF_2 by epitaxy. We have begun a program to do so and this is a report of the initial studies of antiferromagnetic resonance (AFMR) of MnF_2 thin films epitaxially grown on MgF_2, ZnF_2, and FeF_2 substrates. The reasons for choosing the rutile-structured XF_2 materials include the relative ease of preparation of films by evaporation, the close match in the lattice constants,[11] and the considerable understanding that now exists of the magnetic properties of these fundamental systems in the bulk.

Unlike FMR, which is insensitive to variations in the exchange interaction that might arise from strains, imperfections, or surface (interface) contributions, AFMR is extremely sensitive to whatever might contribute to changes in both the exchange and anisotropy interaction. This can be seen by comparing the equations for the uniform mode frequency in the ferromagnet with that of the antiferromagnet[12]

$$\text{FMR: } \omega_0/\gamma = H_0 + (N_\perp - N_z)M \ , \tag{1}$$

$$\text{AFMR: } \omega_0/\gamma = (2H_A H_E + H_A^2 + 2N_\perp M_s H_A)^{1/2} \pm H_0 \ , \tag{2}$$

where H_0 is the external field applied parallel to the c axis, N_z and $N_\perp$ are, respectively, the longitudinal and transverse demagnetizing factors, M and M_s are the magnetization and sublattice magnetization, respectively, H_A the anisotropy field, H_E the exchange field, and γ the gyromagnetic ratio.

This extreme sensitivity of AFMR to what could be large variations in H_E and H_A in deposited films imposes stringent demands on the degree of perfection that must be achieved in the epitaxy process. Indeed this proved to be the case. Only in the best films, as judged by the x-ray texture camera pictures, could any AFMR be observed. However, the AFMR proved itself more sensitive than the texture camera in distinguishing between the quality of the films. Films that were perfect as far as could be judged from texture camera pictures could still be distinguished by their AFMR linewidth.

EXPERIMENTAL METHODS

Careful substrate preparation, including both mechanical and chemical etch polishing, and substrate heating to approximately 600 °C were all found to be necessary to obtain good epitaxy. This was determined to be the case by the use of the x-ray texture camera technique to be described in detail below. A well-prepared epitaxial film produces a texture camera picture with an array of Bragg diffraction spots symmetric with that of the substrate but slightly offset because of the small lattice-spacing mismatch between the film and substrate. Partial epitaxy caused by poor substrate preparation or low substrate temperature resulted in texture camera pictures with polycrystalline lines and/or unexpected Bragg diffraction spots indicating probable growth in other crystalline directions.

A Laue x-ray technique was to orient the 1-cm-diameter, 2-mm-thick MgF_2, ZnF_2, and FeF_2 substrates with the c axis perpendicular to the major face to within

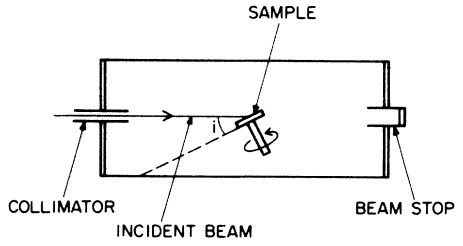

FIG. 1. Diagrammatic sketch of cylindrical texture camera technique; i is the angle of incidence (usually 30°). The film occupies the inner surface of the cylindrical body, while the sample is rotated about the indicated axis.

$\frac{1}{2}$ of a degree. The crystal faces were then ground parallel. The crystals were first mechanically polished in sequential steps with 5-μm, 1.0-μm alumina, and 0.06-μm colloidal silica. To avoid contamination of the polishing solutions, each crystal was carefully cleaned and rinsed between each step. Careful sequential solvent cleaning in trichloroethylene, acetone, ethanol, and distilled H_2O afterward removed any wax residue. Finally, 1:10 dilu-

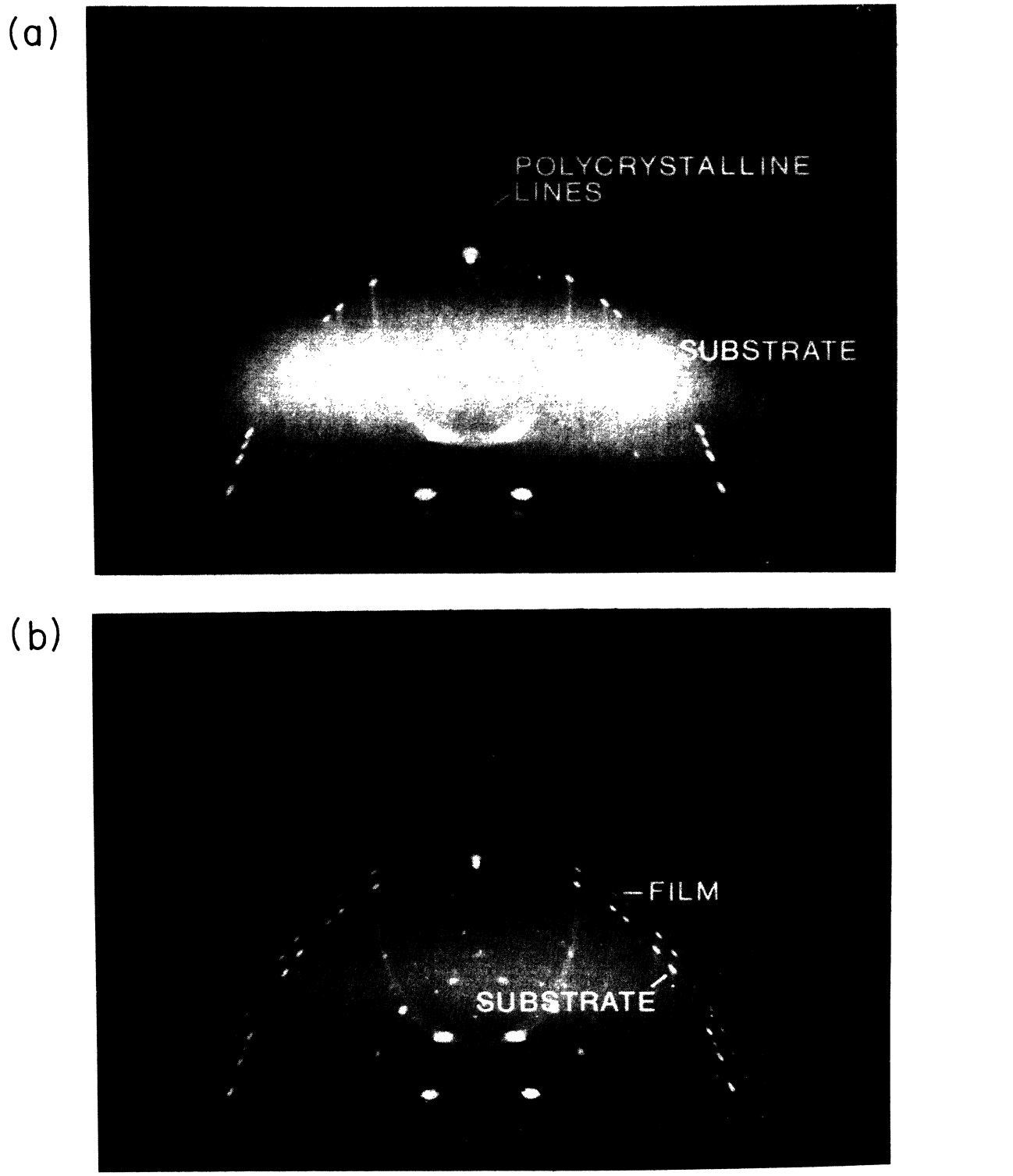

FIG. 2. (a) X-ray texture patterns of a polycrystalline MnF_2 film on ZnF_2, Cu $K\alpha$ radiation. Note the horizontal polycrystalline lines. (b) X-ray texture pattern of epitaxial MnF_2 film on ZnF_2, Cu $K\alpha$ radiation. Note the symmetric diffraction patterns from the MnF_2 and ZnF_2 substrate, indicative of parallel epitaxy. One is slightly offset from the other because of the lattice-constant mismatch.

tion of HCl in distilled H_2O was used as an etch polish, after which the crystal was rinsed in distilled H_2O.

The oriented, polished crystal was placed in a substrate heater mount consisting of a Cu holder and button heater with an attached Chromel-Alumel thermocouple. To avoid hydrocarbon contamination during substrate heating and MnF_2 deposition, high vacuum was attained by roughing to 200 mtorr with a well-trapped mechanical pump, followed by cryopumping to the mid 10^{-6}-torr range. At $\sim 5\times 10^{-6}$ torr, the poppet valve to the ion pumps was opened and the base pressure of mid 10^{-8} torr was reached in approximately 3 h. This pressure was maintained during substrate heating to ~ 600 °C. However, during MnF_2 deposition, the chamber pressure rose to the low 10^{-6}-torr range.

MnF_2 deposition took place by thermal evaporation from heated, pyrolyzed graphite crucibles. Film thicknesses and evaporation rates were measured with a quartz-crystal thickness monitor. Typical evaporation rates were ~ 10 Å/sec and were controlled manually. Film thicknesses ranged from 0.5 to 3 μm. After the deposition, the sample was allowed to cool, then vented to ultrahigh-purity N_2 gas.

The structure and texture of the films have been studied by x-ray diffraction using a cylindrical texture camera[13,14] (see Fig. 1). The diffraction pattern is recorded on a cylindrical photographic film coaxial with the incident beam. The sample is rotated and inclined at a fixed angle with respect to the incident beam. Diffraction from polycrystalline material intersects the recording film in straight lines at the proper 2θ angle. Single-crystal material produces well-defined Bragg diffraction spots. The degree of crystallinity of the material may be determined by observing whether lines or spots appear (see Fig. 2). This technique provides a convenient, quick, and inexpensive method of determining the degree of epitaxy obtained in the thin-film systems. Good epitaxy results in an array of Bragg diffraction spots symmetric with those of the substrate but slightly offset because of the lattice constant mismatch between film and substrate (see Fig. 2).

AFMR was measured with a conventional K-band (22–26 GHz) microwave reflection spectrometer.[12] A phase-locked klystron was coupled to a conventional magic-T bridge and a field-modulation and phase-sensitive detection technique was employed. The sample is mounted near the shorted end of the wave guide at one bridge arm. The dc field H_0 was provided by a 13-T superconducting magnet and was measured with a wide-band NMR magnetometer giving an accuracy of ± 3 Oe. All measurements were carried out at 4.2 K.

EXPERIMENTAL AFMR RESULTS AND DISCUSSION

AFMR in a thick disk of MnF_2 occurs at $\omega/2\pi=260$ GHz at $H=0$ and 4.2 K. The field-decreasing branch [see Eq. (2)] may be brought into resonance at 24 GHz with a field $H_0\simeq 84$ kOe applied parallel to the c axis.[12] In Fig. 3 we show the observed spectrum in a thick disk of MnF_2 which reveals both uniform (AFMR) and magnetostatic modes,[12] the latter arising from the nonellipsoidal shape of the disk. In the epitaxial films only the uniform mode appears to be excited. Shown in Fig. 3 is one of the narrowest resonance lines observed in films of MnF_2 epitaxially deposited onto ZnF_2 substrates; AFMR was observed in a number of other epitaxial films with widths that varied between 15 and 500 Oe. The line profiles in all cases appeared to be Lorentzian in shape.

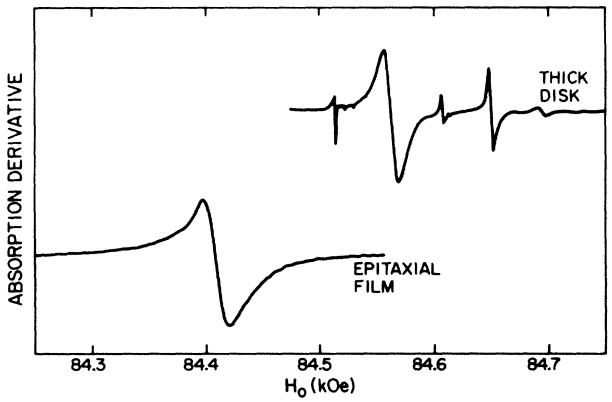

FIG. 3. Comparison of the AFMR spectra of a typical MnF_2 thin film, with a thickness of 2.7 μm, and a MnF_2 thick disk, with a thickness of 3 mm. Note that both the uniform and magnetostatic modes appear for the thick disk, while only the uniform mode appears in the thin film. The resonance position of the thin film appears below the bottom of the magnon band of the bulk MnF_2 thick disk.

There are two points to be made in comparing the resonances in the epitaxial films with that found in the thick disk made from bulk MnF_2; one relates to the relative widths and the other to the relative field at which resonance occurs for the same frequency. For an ideal infinitely thin film, the transverse demagnetizing factor $N_\perp=0$ and the AFMR frequency [see Eq. (2)] is identical with the lowest spin-wave frequency as obtained from the extrapolation to $k=0$ of the spin-wave dispersion[12]

$$\omega_k/\gamma=(H_c^2+2H_E b^2k^2)^{1/2}\pm H_0 \ . \tag{3}$$

Here $H_c^2=2H_AH_E+H_E^2$ and $b=az^{-1/2}$ with a being the nearest-neighbor separation of spins on opposite sublattices and z their number. Thus the uniform mode in the epitaxial film is degenerate only with states very near the bottom of the spin-wave band. At low temperatures ($kT\ll\hbar\omega_0$), where thermal excitation of spin waves is unimportant, the lifetime of the uniform mode is extremely long and would correspond to a linewidth of less than 1 mOe.[15] Only two-magnon "imperfection" scattering should then contribute to the linewidth.[15] But because the uniform mode is degenerate with so few (if any) states in the band, one would expect this process to be ineffective in contributing to the broadening. Indeed we expected to find extremely narrow ($\sim$mOe) linewidths in the epitaxial films and the observation of large linewidths (in some cases in excess of 300 Oe) might seem at first surprising.

However, before considering what other mechanisms might contribute to line broadening, including possible unresolved magnetostatic modes, we note that a correlation exists between the resonance fields H_0 at a fixed frequency and the linewidth. This is seen in Fig. 4, where H_0 versus linewidth is plotted for the different epitaxial

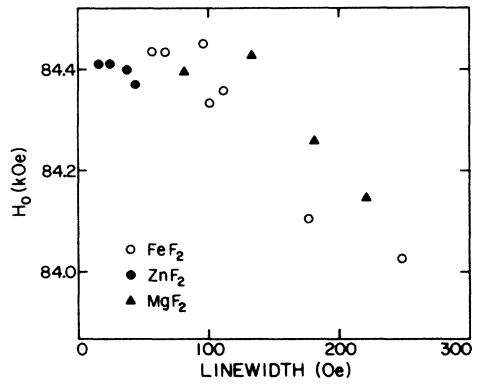

FIG. 4. Resonance position of the uniform mode versus the linewidth of epitaxial MnF_2 films on various substrates. Note that the linewidth monotonically decreases as the resonance field H_0 approaches what we believe to be the bottom of the band in the bulk MnF_2 sample.

MnF_2 films. Note that for those on ZnF_2 or MgF_2 substrates the linewidth monotonically decreases as H_0 approaches what we believe to be the bottom of the band in the bulk MnF_2 sample.[15] The fact that H_0 in the films lies *below* the bottom of the band in the bulk sample suggests that the average value of H_c in the films $\langle H_c \rangle < H_c$ in the bulk. Furthermore since $H_0(\text{bulk}) - H_0(\text{film})$ correlates with the linewidth it would appear that the origin of the linewidth is to be understood in terms of whatever it is that causes $\langle H_c \rangle$ to deviate from H_c in the bulk.

Without further study of the properties of the films (mosaic structure, voids, roughness of the substrate surface, strain fields, and dislocation densities) one can only speculate as to the significance of the H_0-linewidth correlation. However, it is perhaps more important at this early stage to accentuate the *positive* results of these studies; namely, linewidths as narrow as 15 Oe imply a degree of perfection such that the superexchange interaction J has a root-mean-square variation $\Delta J/J \simeq 10^{-4}$ throughout the film. Since J varies roughly as $(r - r_0)^{-12}$, this implies the root-mean-square variation in the lattice spacing throughout the film is no larger than $\Delta r_0/r_0 \simeq 10^{-5}$. Thus a film of this quality should give a broadening of the second-order phase transition of no more than $\Delta T_N/T_N \simeq 10^{-4}$, which would be quite adequate for critical phenomena studies.

Obviously further studies, including independent characterization methods, are called for before the line broadening mechanism can be understood. These are being undertaken at the present time in conjunction with the first efforts to produce multilayer films of these same materials (e.g., MnF_2-FeF_2 layers on ZnF_2).

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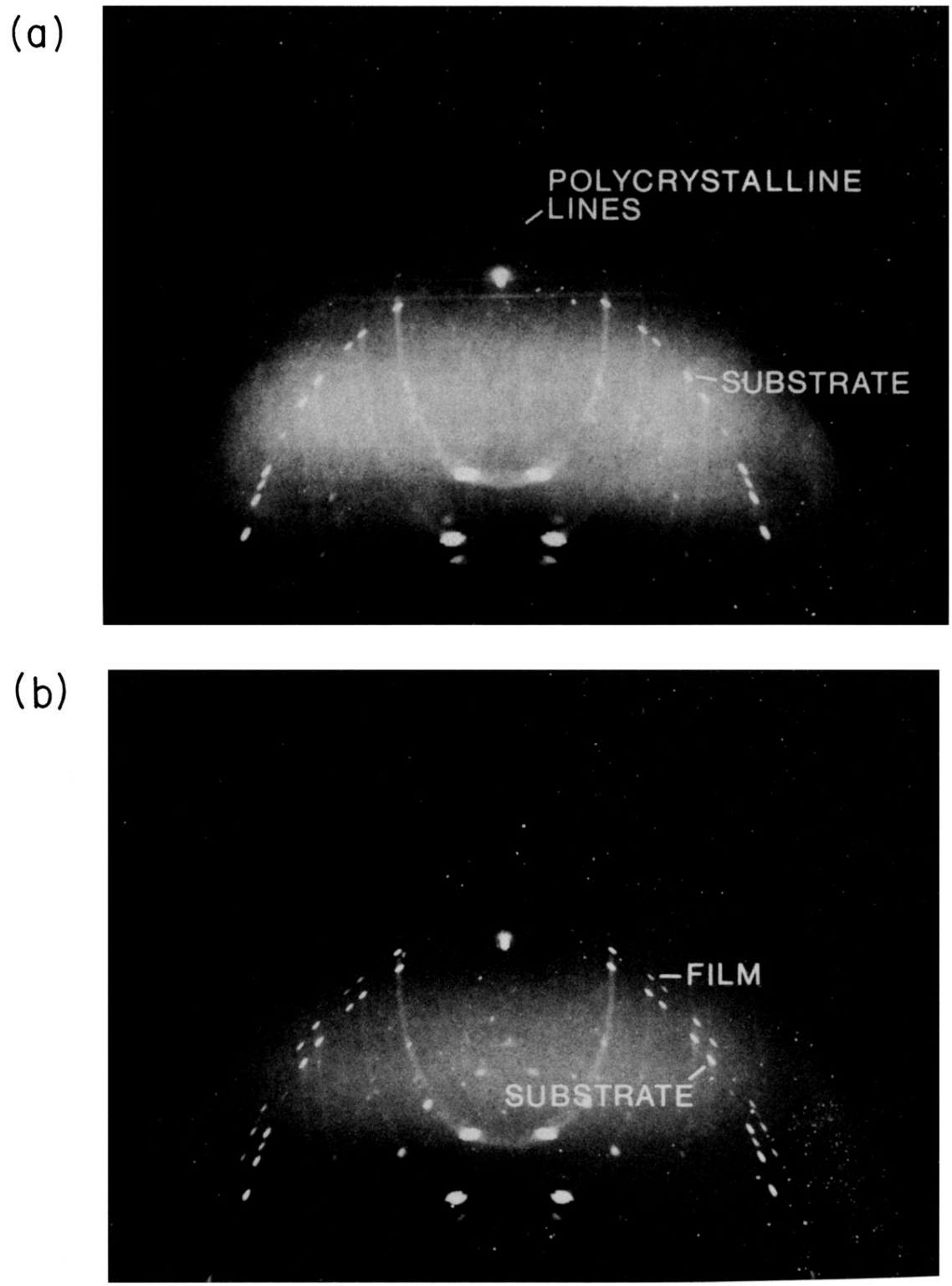

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