(now returned).
[1]*Structural Phase Transitions and Soft Modes*, edited by E. J. Samuelson and J. Feder (Universitetsforlaget, Oslo, Norway, 1971), and references quoted therein.
[2]K. A. Müller, W. Berlinger, and F. Waldner, Phys. Rev. Letters 21, 814 (1968).
[3]K. A. Müller and W. Berlinger, Phys. Rev. Letters 26, 13 (1971).
[4]T. Riste, E. J. Samuelson, K. Otnes, and J. Feder, Solid State Commun. 9, 1455 (1971).
[5]G. Shirane, V. J. Minkiewicz, and A. Linz, Solid State Commun. 8, 1941 (1970).
[6]V. Plakhty and W. Cochran, Phys. Status Solidi 29, K81 (1968); W. J. Minkiewicz, Y. Fujii, and Y. Yamada, J. Phys. Soc. Japan 28, 443 (1970).
[7]E. Courtens (unpublished).
[8]F. Borsa, in Ref. 1.
[9]F. Borsa, M. L. Crippa, and B. Derighetti, Phys. Letters 34A, 5 (1971).
[10]G. Bonera, F. Borsa, M. L. Crippa, and A. Rigamonti, Phys. Rev. B 4, 52 (1971).
[11]M. Furukawa, Y. Fujimori, and H. Jirakawa, J. Phys. Soc. Japan 29, 1528 (1970).
[12]K. Gesi, J. D. Axe, G. Shirane, and A. Linz, Phys. Rev. B 5, 1933 (1972).
[13]A. Okazaki and Y. Suemune, J. Phys. Soc. Japan 16, 671 (1961).
[14]K. Hirakawa, K. Hirakawa, and T. Hashimoto, J. Phys. Soc. Japan 15, 2063 (1960).
[15]M. B. Walker and R. W. H. Stevenson, Proc. Phys. Soc. (London) 87, 35 (1966).
[16]R. M. Sternheimer, Phys. Rev. 146, 140 (1966).
[17]In $LaAlO_3$ there is direct experimental evidence that the measured efg is proportional to the square of the rotational displacement parameter (see Ref. 9).
[18]R. Comes, F. Denoyer, L. Deschamps, and M. Lambert, Phys. Letters 34A, 65 (1971).
[19]Th. von Waldkirch, K. A. Müller, W. Berlinger, and H. Thomas, Phys. Rev. Letters 28, 503 (1972).
[20]G. Bonera, F. Borsa, and A. Rigamonti, J. Phys. (Paris) 2, C195 (1972).

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Possibility of Guided-Neutron-Wave Propagation in Thin Films

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The propagation of guided-neutron waves in thin films is discussed and a theory is given for a simple coupling device for exciting such guided-neutron waves by means of neutron beams. The principle is analogous to that used in the construction of optical-prism couplers used to guide laser beams in thin films. The larger divergence of the incident neutron beam drastically lowers the effective coupling efficiency, but the effect should nevertheless be readily observable. A qualitative discussion of Marx's microguide device and its relationship to our problem is also given.

The phenomenon of total internal reflection of neutron beams is well known[1] and has been successfully used for the construction of macroscopic neutron guide tubes situated at nuclear reactors.[2] The effect is completely analogous to the corresponding optical phenomenon, and may be explained in terms of the effective refractive index for neutrons which is given by

$$n^2 = 1 - V/E_0 \,, \tag{1}$$

where V is the average potential experienced by the neutron of energy E_0 in the medium and is given by

$$V = 2\pi\hbar^2 Nb/m \,, \tag{2}$$

where m is the neutron mass, b is the coherent scattering length, and N is the number of atoms per unit volume. [Equations (1) and (2) are only valid provided Bragg reflections do not occur and other scattering and absorption processes are negligible.] Recently, much interest in optics has centered on the properties of thin-film optical waveguides,[3] in which a thin film is surrounded by media of lower refractive indices and is capable of sustaining a discrete set of guided modes, each composed of two coherent beams undergoing successive total internal reflections at each interface. Such guided modes should also be possible for neutrons propagating in a thin film surrounded by media of lower refractive index.

The purpose of this paper is to discuss the coupling of the neutron beam by means of the evanescent field associated with total internal reflection at the air-medium interface. This method is completely analogous to the prism coupler in optics where it has been shown that conversion efficiencies of 80% can be realized.[4,5] There are a number of reasons peculiar to neutron technology which makes this high efficiency not realizable, as will be discussed later.

Recently, Marx[6] has reported a technique for using bent thin-film sandwiches to deflect neutron beams through wide angles over short distances, and has reported 97% efficiencies for the neutrons emerging from the end of the device after undergoing several total internal reflections. There are several differences between Marx's technique and the method described here. These pertain to the spatial confinement of the guided neutron beam and to the momentum resolution attainable with the device in a direction normal to the film. We should emphasize that the purpose of the method described here is not to efficiently deflect neutron beams using small radii of curvature, as in Marx's experiment, but rather to achieve the true guided modes intrinsic to a thin film with the corresponding beam confinement and momentum resolution, with, of course, a consequent large reduction in final neutron intensities. A more detailed analysis of the two methods is given later in this paper.

A schematic diagram of the coupler is shown in Fig. 1(a). The incident neutron beam is incident at an angle θ_0 less than the critical angle for medium 2. Normally such a beam would be totally reflected and there would simply be an exponentially decreasing wave in medium 2. However, if the component of the neutron wave vector parallel to the film k_x is exactly matched to that of one of the resonant guided modes possible in the film (medium 3) for that particular energy of the neutron, the exponentially *decreasing* wave in medium 2 becomes an exponentially *increasing* wave and builds up to a very large amplitude in medium 3, corresponding to a neutron tunneling through the border region 2 and being resonantly trapped in region 3. Owing to the finite thickness of medium 2 [in the region AB in Fig. 1(a)], the trapped particles would eventually leak out again and be totally reflected back into medium 1. This may be thought of as a broadening and perturbation of the resonance due to the presence of medium 1. Thus, if we consider a finite beam impinging on medium 2 as shown, the flux of trapped particles in film 3 would build up to a maximum and then rapidly decrease beyond point B owing to leakage. At point B, however, medium 2 is made very thick so that the flux stays trapped in the film. One may think of the phenomenon as a *displacement* of a totally reflected beam from the first interface induced by the resonant modes in the film. If the displace-

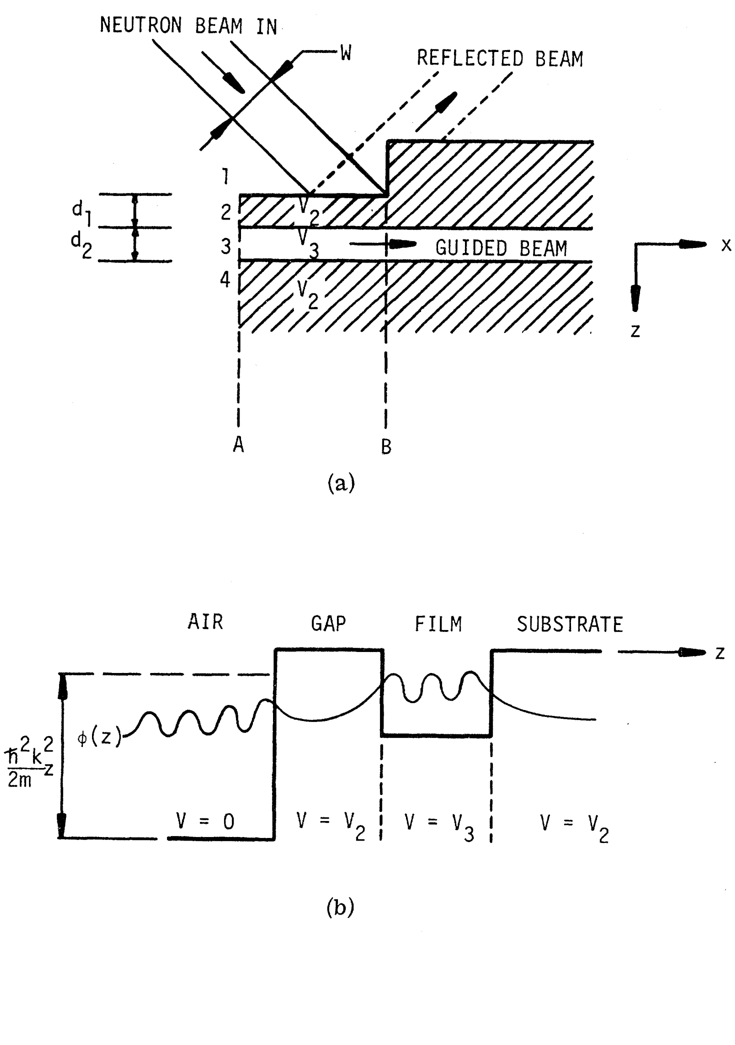

FIG. 1. (a) Schematic diagram of neutron-beam thin-film coupler. (b) Schematic diagram of the effective potential variation in the z direction and a sketch of the neutron wave function at conditions for resonance with a guided mode.

ment is comparable to the beam width, the beam may be trapped in the film by making medium 2 thick. The principle is similar to that of the Goos-Hänchen effect in optics.[7] For optimum coupling into the film the thickness of medium 2 in region AB depends on several factors including the beam width W, as shown below. The guided beam in the film may then be extracted by making medium 2 thin again on the other side of the sample, as shown, or it may be allowed to exit through the end of the film. This physical description also applies to the prism coupler in optics, and a one-to-one correspondence between the potential and the electromagnetic problem can be made for transverse electric polarization, i.e., TE modes. In the electromagnetic case, region 1 (AB) is the prism, region 2 is the air gap, region 3 is the waveguide, and region 4 is the substrate.

To calculate the efficiency of the coupler illustrated in Fig. 1 we follow the procedure used in optics, namely, allowing region AB to be infinite in extent and calculate the efficiency at the point B in the medium for a finite incident beam of width W. This simplification makes the problem separable in the x and z directions and leads in the z direction to a variation of the familiar square-well-potential problem illustrated in Fig. 1(b).

In Fig. 1(b), k_z is the transverse wave number. Taking the wave function to be $\phi(z)e^{ik_x x}$, the one-dimensional problem becomes

$$\frac{d^2}{dz^2}\,\phi_j(z) + (k_z^2 - k_{v,j}^2)\phi_j(z) = 0\ , \tag{3}$$

where $k_{v,j}^2 = (2m/\hbar^2)V_j$ and the index j denotes the layer. For the electromagnetic case, $\phi_j(z)$ is the variation in the z direction of the y component of the electric field and $k_{v,j}^2 \rightarrow k_0^2(1-\chi_j)$, where χ_j is the dielectric constant in medium j. Recall that for TE polarization, $\vec{E} = (E_x, E_y, E_z) = (0, E_y, 0)$.

In the final analysis it will be shown that the maximum coupling occurs when $\Gamma W \simeq 1$, where Γ is the width of the bound modes. Γ is controlled by d_1, since if d_1 is infinite the amplitude cannot leak out. The above criterion is intuitively reasonable because W controls the spread in k_z and Γ controls the spread in the resonance. The distinction between the neutron problem and the laser problem is that neutron beams have a much larger spread in k_z arising from the noncoherence of neutron beams. It is important to note that the first experiment with the prism coupler was performed with white light.[3]

Let us now return to the neutron case and analytically illustrate these points. The wave function in the various layers can be written

$$\psi_j(x,z) = [1/(2\pi)^{1/2}] \int_{-\infty}^{+\infty} \exp[i(k\sin\theta_0 + k_M\cos\theta_0)x] \times \phi_{k,j}(z)\,dk\ , \tag{4}$$

where $k_M^2 + k^2 = k_0^2$. The amplitudes $\phi_{k,j}(z)$ are taken to correspond to a linear superposition of the characteristic solutions in the respective layers. As illustrated in Fig. 1(b) we have taken the material in medium 4 to be the same as in region 2. In addition, we suppose the condition $V_3 < \hbar^2 k_1^2/2m < V_2$ is satisfied. This assures that the z component of the wave is propagating in regions 1 and 3 but nonpropagating in regions 2 and 4. From the differential equation the propagation constants in the z direction in the various media are given by

$$\begin{aligned} k_1 &= k_M\sin\theta_0 - k\cos\theta_0\ ,\\ k_2 &= (k_{v,2}^2 - k_1^2)^{1/2}\ ,\\ k_3 &= (k_1^2 - k_{v,3}^2)^{1/2}\ . \end{aligned} \tag{5}$$

Applying the usual boundary conditions at the interfaces of the various regions leads to six homogeneous equations relating a total of seven amplitudes. These amplitudes can be expressed in terms of the Fourier component of the incident wave. If we take the incident beam of width W to be a step function as illustrated in Fig. 1(a), then we have all the amplitudes proportional to $[W/(2\pi)^{1/2}][(\sin\frac{1}{2}kW)/\frac{1}{2}kW]$. Defining the total efficiency $\eta_m(x)$ for coupling into the mth mode as the ratio of the total particle current in the x direction in regions 2–4 to the total incoming particle current $I_i = [(\hbar/m)k_0]W$, we obtain

$$\eta_m(x) \simeq Q(1 - \exp\{-\Gamma_m[x\tan\theta_0 + \tfrac{1}{2}W(\sec\theta_0)]\})^2\ ,\quad -W/2\sin\theta_0 \leq x \leq W/2\sin\theta_0 \tag{6}$$

$$\simeq Qe^{-2\Gamma_m\tan\theta_0 x}\{4\sinh^2[(\tfrac{1}{2}\Gamma_m W)\sec\theta_0]\}\ ,\quad x \geq W/2\sin\theta_0\ , \tag{7}$$

where

$$Q = 2\Gamma_m\cos\theta_0\{\{(k_3^m)^2(1-e^{-2k_2^m d_1}) + \{2(k_2^m)^2 + [(k_2^m)^2 + (k_3^m)^2]k_2^m d_2\} + (k_3^m)^2\}\} \times \{W[\Gamma_m^2 + (k_0\sin\theta_0 - k_{1s}^m)^2][(k_2^m)^2 + (k_3^m)^2] \times (2 + k_2^m d_2)\}^{-1}\ , \tag{8}$$

$$k_{1s}^{(m)} = k_1^m + \Delta_m\ ,$$

$$\Delta_m = \frac{2[(k_1^m)^2 - (k_2^m)^2](k_2^m k_3^m)^2 e^{-2k_2^m d_1}}{k_1^m[(k_1^m)^2 + (k_2^m)^2](2 + k_2^m d_2)[(k_2^m)^2 + (k_3^m)^2]}\ , \tag{9}$$

$$\Gamma_m = \frac{4k_2^m(k_2^m k_3^m)^2 e^{-2k_2^m d_1}}{[(k_1^m)^2 + (k_2^m)^2](2 + k_2^m d_2)[(k_2^m)^2 + (k_3^m)^2]}\ . \tag{10}$$

The values of k_1^m occurring in the above equations are obtained from the solutions of the following transcendental equations:

$\tan\frac{1}{2}k_3 d_2 = k_2/k_3$, even modes

$\tan\frac{1}{2}k_3 d_2 = -k_3/k_2$, odd modes.

Also, k_2^m and k_3^m are evaluated with $k_1 = k_1^m$. The quantities Δ_m and Γ_m are the shift and linewidth of the bound states of the square-well potential due to the finite dimension of region d_1. Note that if $d_1 \to \infty$, both Δ_m and $\Gamma_m \to 0$. The above expression for $n_m(\chi)$ was obtained by first-order expansion about k_1^m, i.e., $k_1 = k_1^m + \Delta k$ and taking $k_0 \sin\theta_0 \simeq k_1^m$. The three terms in the double curly brackets of the numerator of Eq. (8) represent the contributions of regions 2, 3, and 4, respectively. In general, the contribution of region 3 will dominate. The maximum coupling efficiency will occur at $x_0 = W/2\sin\theta_0$, i.e., at the edge of the illuminated region. For larger x, the particle current in the film will decay rapidly due to leakage back to medium 1, but this may be avoided by making region 2 very thick for $x \geq x_0$, thus trapping the particles in the film, apart from small losses due to scattering because of the discontinuous boundary condition at $x = x_0$. By Fourier decomposing the finite incident beam and Fourier resynthesizing the beams in regions 2–4, we see that, near a resonance, an effective compression of the incident-beam current into regions 2–4 has been obtained. For an incident beam of given θ_0 and k_0, this compression may be considerable; an examination of the expression for $\eta_{\max}$ shows that it can be as large as 0.81 when $k_0 \sin\theta_0 = k_1^m$. This is achieved when $\Gamma_m W \sec\theta_0 \simeq 1.26$. If Γ_m is very much greater than the optimum value, the leakage rate becomes too large and a sufficiently large current cannot be built up in the film over the illuminated region. On the other hand, if the resonance is too sharp (Γ_m too small) the particles cannot tunnel across to region 3 with sufficient probability for the current to build up rapidly enough.

Because neutron beams, in contrast to laser beams, are by no means monochromatic or well collimated on the scale of the widths of the resonances, we have to consider a neutron beam possessing finite instrumental resolution. Each guided mode in the film acts to select out for channeling only a very small bandwidth of neutrons for wave vectors normal to the film, but all possible guided modes are excited by the finite-resolution incident beam. Even though the effective potential barriers in the neutron case are very small, a considerable number of such guided modes may still exist in a thin film.

To illustrate the effects we have considered a 1000-Å-thick film of aluminum ($4\pi Nb = 0.27 \times 10^{12}$ cm^{-2}) surrounded by nickel ($4\pi Nb = 1.19 \times 10^{12}$ cm^{-2}), in which there turns out to be four possible guided modes. A 3-Å 0.1-mm-wide neutron beam of angular divergence $\pm 0.5°$ incident on a 200-Å-thick nickel epilayer at a mean θ_0 of $0.22°$ would have a fraction 4×10^{-4} of the neutrons trapped in the guided modes. This is a number which is well within detectable levels with present day fluxes. Although such fluxes are still sufficiently low that it does not seem possible to actually perform neutron scattering studies of the thin films using these guided beams, the above technique seems to be a fairly elegant and convenient method of arranging for a neutron beam to propagate purely in a film and avoid the bulk material. An alternative method of guiding neutrons in the film is by direct entry through the end of the film at a grazing angle less than the critical angle. However, this method is likely to be less convenient in practice for the following reasons: (a) the necessity of masking off the bulk part of the sample from the incident neutron beam, and (b) for very thin films, diffraction effects at the entry slit will cause considerable losses in the guided beam. This is so even if the film thickness is ~100 neutron wavelengths, since the "potential barrier" for the guided neutron beams is extremely weak. Thus in the numerical example given above only that fraction of the finite incident beam which is resolvable into the four possible guided modes will propagate. In this connection, we should point out that Marx's experiment does not correspond to excitations of discrete guided modes in a *single* thin film as discussed above. This is because the alternating layers of nickel and aluminum which are used in the microguide in his experiment result not in the single square-well potential illustrated in Fig. 1(b) but rather in a periodic array of such square-well potentials separated by thin- and weak-barrier regions. This periodic array finally has macroscopic dimensions. As is well known from the theory of the Kronig–Penney model for electrons, of which the present case is the exact analog, the discrete (degenerate) bound states in each square well individually are now replaced by a set of bands which, in the limit that the array becomes infinite, merges into a continuum, except for possible small forbidden regions due to the "zone gaps." Thus a neutron which enters the end of the guide will find not simply a discrete set but a semicontinuum of states which correspond to "guided modes" in the *microguide as a whole*. The neutron beams are now no longer confined to one thin film but tunnel back and forth between the potential wells. Since the neutron is not here confined to a discrete resonance in a single film, the momentum resolution in the perpendicular direction will roughly correspond to the momentum resolution in that direction of the incident beam, i.e., typically ~10^6 cm^{-1}. We assume here that the bending of the microguide is a macroscopic phenomenon, so that the above picture of quasicontinuous states stays true but the mean direction of the x axis slowly changes as the microguide is traversed.

In contrast, one of the novel features of the present technique for launching neutron waves in thin films is the high-momentum resolution achievable in the z direction. Since we are dealing with discrete resonances, this resolution is determined purely by the width of a given resonance Γ_m, which in turn is determined by the width of the gap region d_1. For the numerical example discussed in this paper (namely, a 1000-Å-thick film of aluminum with a 200-Å nickel epilayer) the width of the resonance for optimum coupling turns out to be ~100 cm^{-1}, compared to a k_0 of ~10^8 cm^{-1}. A convenient way to get neutrons with this momentum resolution out of the guide then would be to make the nickel epilayer thin again (~200 Å) at the other end of the guide. The reverse coupling process will then take place and the neutrons will emerge smoothly from the guide, corresponding to the exponential decay of particle current in the film as given by Eq. (6). If only one mode is desired it may be achieved by choosing a film which is thin enough to contain only one mode, or also the emerging beam may then be allowed to impinge on a second guide only one of whose modes coincides with the mode desired. Since in a given mode the coupling efficiency is ~80%, no further significant losses are anticipated using such an "analyzer" technique. The method proposed here thus describes a way of producing extremely highly collimated neutron beams over small distances and such beams could find several applications in neutron physics experiments, such as the possibility of shifting the resonance by applying external fields to the guide. Such experiments then would provide a very sensitive test for the interaction of the neutrons with such external fields. We have neglected here the inevitable loss of resolution due to nonhomogeneous thicknesses of the films, but we believe that these are not an inseparable obstacle to attaining the required resolution.

It is hoped that the analysis presented in this paper provides the stimulus for experimentation in what appears to be a novel application of neutron physics.

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[1]G. Bacon, *Neutron Diffraction*, 2nd ed. (Clarendon, Oxford, England, 1962), p. 116.

[2]B. Farnoux, B. Hennion, and J. Fagot, *Neutron Inelastic Scattering* (International Atomic Energy Agency, Vienna, 1968), Vol. II, p. 353; H. Maier-Leibnitz and T. Springer, J. Nucl. Energy A/B **17**, 217 (1963).

[3]J. Kane and H. Osterberg, J. Opt. Soc. Am. **54**, 347 (1964); E. R. Schineller, R. F. Flam, and D. W. Wilmot, J. Opt. Soc. Am. **58**, 1171 (1968).

[4]P. K. Tien and R. Ulrich, J. Opt. Soc. Am. **60**, 1325 (1970).

[5]J. E. Midwinter, IEEE J. Quantum Electron. **6**, 583 (1970).

[6]D. Marx, Nucl. Instrum. Methods **94**, 533 (1971).

[7]H. K. V. Lotsch, Optik (Stuttg.) **32**, 116 (1970).

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Mössbauer Line Shift in Iron

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The Mössbauer line shift of iron in FeF_3 is reexamined from the point of view of its temperature dependence. The time-averaged correction to the isomer shift arising from the virtual excitation of an electron from $3d^5$ to $4s$ with the simultaneous emission or absorption of one phonon is calculated and compared with recent experiments.

Recently, Perkins and Hazony[1] examined the temperature dependence of Mössbauer resonance energy as the sum of the second-order Doppler shift and the isomer shift. These workers very carefully examined the experimental data separating the effect of thermal expansion (implicit) and that of the harmonic lattice vibrations (explicit) in several compounds and found that in the case of FeF_3 there is an explicit temperature dependence in the isomer shift as previously predicted[2] by the present author. In the present paper, it is proposed that the admixtures of the s-like electronic configurations by the electron-phonon interaction produce corrections to the isomer shift. Taking the example of $Fe^{3+}(3d^5)$ in FeF_3, the corrections to the isomer shift owing to the admixtures of the crystal field states $\Gamma_3(3d^4 4s)$ and $\Gamma_5(3d^4 4s)$ into the ground-state configuration are calculated and the results compared with experiments.

The electron-phonon interaction arising from the modulation of the octahedral crystal field by lattice phonon is[3]

$$\mathcal{H} = \sum_{j=2}^{6} V_j Q_j , \tag{1}$$