by inelastic scattering of low-energy electrons. Similar results (although corresponding to other r_s values) should be obtainable with any substrate plus nearly-free-electron metal film which exhibits a minimum in $\Phi(N_S)$.

We are grateful to Conyers Herring for pointing out the connection between this work and that of Smoluchowski, Ref. 8.

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*Karlsruhe University, Karlsruhe, Germany.

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PHONON-PUMPED NUCLEAR SPIN WAVES IN A FLOPPED ANTIFERROMAGNET*

A. Platzker and F. R. Morgenthaler
Department of Electrical Engineering, and Center for Materials Science and Engineering,
Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

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Phonon-exchange pumping of nuclear magnon pairs in low-anisotropy flopped antiferromagnets is proposed and the effective threshold fields given. Although the electronic sublattice magnetizations are driven, magnetoelastically, far below their resonance frequency, the lower pump frequency compensates for the weakened dynamic coupling between nuclear and electronic magnons. The required phonon power density is thus low and well within experimental reach.

When a sufficiently intense elastic wave of the proper frequency propagates through an antiferromagnet, spin-wave pairs can be driven unstable because of coupling via the linear magnetostriction. This method of excitation – related to but different from photon exchange pumping[1] – is of special interest because the sign of the torque on each magnetic sublattice is opposite, giving rise to an "optical-like" excitation that brings the intersublattice coupling into play. Morgenthaler[2] has previously derived the thresholds for phonon parallel pumping of electronic magnon pairs in the unflopped state with special attention to $RbMnF_3$. Although the thresholds are predicted to be low, the pump frequency must be comparable with twice the electronic resonances frequencies – which for $RbMnF_3$ are typically in the 10-GHz range. Unfortunately, ultrasonic transducer fabrication in this range is not within the current state of the art; this fact has inhibited our experimental efforts to date. However, because the Mn^{55} ions carry a nuclear magnetic moment, the possibility exists of phonon pumping nuclear magnon pairs (or one nuclear and one electronic magnon) at a much lower frequency.

In this Letter we give the theoretical first-order instability thresholds for such processes in a flopped cubic antiferromagnet for the case of a longitudinal phonon propagating parallel to the equilibrium direction of the sublattice magnetizations. The threshold of purely electronic magnon pairs is included to allow comparisons. In all three cases the excitation is via the exchange interaction between the electronic sublattice magnetizations and the pumping is thus maximally efficient.[1] The threshold elastic powers required (roughly of the same order in the three situations) appear to be very low for antiferromagnets with weak anisotropy and reasonable magnon losses. The leading candidate appears to be $RbMnF_3$.

It has been shown by de Gennes et al.[3] that nuclear spin-wave branches exist due to the Suhl-Nakamura indirect interaction. The possibility of excitation of nuclear magnon pairs by parallel pumping with an rf field (photon parallel pumping) has also been considered by them but the calculated threshold was found to be very high. That process is not an "exchange pumping" but rather pumping via the weak dipole-dipole coupling which creates small ellipticity of spin-wave precession. Ninio and Keffer[4] have considered the simultaneous excitation of one electronic and one nuclear spin wave by photon parallel pumping of an unflopped antiferromagnet. Although the calculated threshold for RbMnF_3 is within experimental reach, the unflopped configuration has not been observed for RbMnF_3 and seems to be unattainable.[5] Hinderks and Richards[6] considered the spin-wave excitation in the flopped state by an rf field applied perpendicular to the magnetizations and parallel to the dc magnetic field[7] and obtained the thresholds for the various processes. Only magnon pairs which are degenerate in $\vec{k}$ can be excited with photon fields since energy and momentum have to be conserved and the photon momentum is negligible.

We assume that a longitudinal elastic displacement

$$\vec{p}=\vec{p}_0\cos(\omega_p t-\vec{k}_p\cdot\vec{r}) \tag{1}$$

(generated through piezoelectric transducers) propagates parallel to the direction of the equilibrium magnetization with a velocity v_1. This wave generates an effective field, through the magnetoelastic interaction, which is assumed to act directly on the electronic sublattice magnetizations and only through the latter, indirectly, on the nuclear magnetization vectors. The direct nuclear-phonon interaction is assumed to be small and hence is neglected. The nuclear magnetization vectors are thus coupled only indirectly through a Suhl-Nakamura effective interaction. The magnetization is assumed to consist of a pair of traveling nondegenerate spin waves together with a dc component.

The equations of motion of the two spin waves are

$$\begin{aligned}\dot{\vec{m}}_{k_1}e^{i\vec{k}_1\cdot\vec{r}}&=A_{k_1}\vec{m}_{k_1}e^{i\vec{k}_1\cdot\vec{r}}+P\vec{m}_{-k_2}e^{-i\vec{k}_2\cdot\vec{r}},\\ \dot{\vec{m}}_{-k_2}e^{-i\vec{k}_2\cdot\vec{r}}&=A_{-k_2}\vec{m}_{-k_2}e^{-i\vec{k}_2\cdot\vec{r}}\\ &\qquad+P\vec{m}_{k_1}e^{i\vec{k}_1\cdot\vec{r}},\end{aligned} \tag{2}$$

where $\vec{m}_{k_1}$ and $\vec{m}_{-k_2}$ are the magnetization vectors with wave vectors $\vec{k}_1$ and $\vec{k}_2$ of amplitudes k_1 and k_2, A_{k_1} and A_{-k_2} are the linear resonance matrices of the spin-wave spectrum, and P is the effective pump matrix.

The magnetization vectors include the electronic and nuclear transverse components of the two sublattices and therefore lead to eighth-order vector equations. A_k includes contributions from the Zeeman, anisotropy, and exchange energies. These are the dominant terms for an antiferromagnet within the frame of the molecular field approximation. We specialize A_k to the case of a flopped cubic antiferromagnet although the formulation of Eq. (2) is general. By first diagonalizing the A_k matrices (transformation into normal modes which gives the four spin-wave branches) and then using perturbation techniques,[8] the threshold for the three above-mentioned processes are calculated. The losses are accounted for phenomenologically by introducing relaxation frequencies $\eta_e(\Omega_k,k)$ and $\eta_n(\omega_k,k)$ for the electronic and nuclear spin-wave branches, respectively.

The magnon pair which is thus "exchanged pumped" by the magnetoelastic wave has to satisfy the energy and momentum conservation conditions

$$\omega_p=\omega_{k_1}+\omega_{k_2},\quad \vec{k}_p=\vec{k}_1+\vec{k}_2. \tag{3}$$

In the flopped state, if the two pumped magnons are of the same kind (both electronic or nuclear), they belong to the same spin-wave branch, contrary to the unflopped state where the pumping is across the gap. Magnons of different kind are pumped only between associated electronic and nuclear branches.

The effective threshold fields for the three processes are given in Table I, where $\omega_{\text{ex}}=\mu_0\gamma_e H_E$ is the exchange frequency; ω_{HF} is the unpulled Mn^{55} nuclear frequency; $\omega_N=\mu_0\gamma_e H_N$, where H_N is the hyperfine field acting on the electronic sublattices; and Ω_{ik} and ω_{ik} are the electronic and nuclear spin-wave frequencies, respectively.

The spin-wave frequencies for a flopped cubic antiferromagnet are

$$\sum_{i=1,2}\Omega_{ik}=[\Omega_{i0}^2+2\lambda\omega_{\text{ex}}\omega_M k^2]^{1/2}, \tag{4}$$

$$\sum_{i=1,2}\omega_{ik}=\omega_{HF}\left[1-\frac{2\omega_{\text{ex}}\omega_N}{\Omega_{ik}^2}\right]^{1/2}, \tag{5}$$

where λ is the effective exchange constant and $\omega_M=\mu_0\gamma_e M$, M being the electronic sublattice magnetization. Ω_{i0} and ω_{i0} are the uniform ($k=0$) antiferromagnetic electronic and nuclear resonances which were given by Freiser et al.[9] and de Gennes,[3] respectively.

Table I. Effective threshold fields for the phonon parallel pumping of various magnon pairs in a flopped antiferromagnet.

Process	Phonon frequency and momentum	Effective threshold field, h_c^{eff}
Excitation of an electronic magnon pair of frequencies Ω_{ik_1} and Ω_{ik_2} $i=1,2$	$\vec{k}_p=\vec{k}_1+\vec{k}_2$ $\omega_{pi}=\Omega_{ik_1}+\Omega_{ik_2}$	$\dfrac{2\left[\eta_e(\Omega_{ik_1},k_1)\,\eta_e(\Omega_{ik_2},k_2)\right]^{1/2}\left[\Omega_{ik_1}\Omega_{ik_2}\right]^{1/2}}{\mu_0\,\gamma_e\,\omega_{ex}}$
Excitation of one electronic and one nuclear magnon of frequencies Ω_{ik_1} and ω_{ik_2} $i=1,2$	$\vec{k}_p=\vec{k}_1+\vec{k}_2$ $\omega_{pi}=\Omega_{ik_1}+\omega_{ik_2}$	$\dfrac{\left[\eta_e(\Omega_{ik_1},k_1)\,\eta_n(\omega_{ik_2},k_2)\right]^{1/2}\left[2\Omega_{ik_1}\omega_{ik_2}\right]^{1/2}\Omega_{ik_1}\Omega_{ik_2}}{\mu_0\,\gamma_e\,\omega_{ex}\,\omega_{HF}\left[\omega_{ex}\,\omega_N\right]^{1/2}}$
Excitation of a nuclear magnon pair of frequencies ω_{ik_1} and ω_{ik_2} $i=1,2$	$\vec{k}_p=\vec{k}_1+\vec{k}_2$ $\omega_{pi}=\omega_{ik_1}+\omega_{ik_2}$	$\dfrac{\left[\eta_n(\omega_{ik_1},k_1)\,\eta_n(\omega_{ik_2},k_2)\right]^{1/2}\left[\omega_{ik_1}\omega_{ik_2}\right]^{1/2}\Omega_{ik_1}^2\Omega_{ik_2}^2}{\mu_0\,\gamma_e\,\omega_{ex}^2\,\omega_{HF}^2\,\omega_N}$

Extensive studies of both the electronic and nuclear $k=0$ modes for the dc magnetic field H_{dc} applied in various crystallographic directions have been recently carried out in this laboratory.[10] At 4.2°K, $\omega_{HF}\sim 686$ MHz and the nuclear modes lie in the uhf region with lowest observed frequency of 280 MHz. This frequency occurs at the spin flop point when a field of ~2400 Oe is applied along a (100) axis. Thus for wave numbers less than 10^6 cm^{-1}, excitation of a nuclear magnon pair should be possible in the frequency range 560-1300 MHz. The electronic modes lie in a much higher frequency range with lowest observed frequency of 5.5 GHz for the same conditions of H_{dc} as above. Thus excitations of an electronic magnon pair should occur at the frequency range 11-20 GHz and one electronic and one nuclear magnon in the range 6-11 GHz. At elevated temperatures the low-frequency limit for the excitation of an electronic pair should be significantly lower, as low as 7 GHz. For the pumping of a nuclear magnon pair, raising the temperature increases the threshold because of the $1/T$ dependence of ω_N and because of the increased losses, and it is not believed that nuclear magnons can be observed above ~10°K.

The threshold power density for a traveling phonon wave is given by

$$P=\tfrac{1}{2}cv_1(k_p\rho_0)^2, \tag{6}$$

where $k_p\rho_0$ is the critical strain which is related to the effective phonon threshold field h_c^{eff}:

$$k_p\rho_0=\mu_0 M h_c^{\text{eff}}/b. \tag{7}$$

b and c are the appropriate magnetoelastic and elastic constants, respectively.

Using published data[11,12] and assuming for the relaxation frequencies those values which give rise to the full observed antiferromagnetic-resonance linewidth, the theoretical threshold power density has been calculated from Table I and formulas (6) and (7). For a wide range of H_{dc}, phonon pump frequency, and temperature, the threshold power density is between 100 and 1000 mW/cm^2, values which can be easily obtained by using piezoelectric transducers. Although attenuation measurements of sound wave propagation at high frequencies (500 MHz and above) in RbMnF$_3$ have not been made, it is not expected to be so high as to prevent the excitation of magnon pairs discussed in this Letter. Phonon parallel pumping experiments (in the sense specified in Ref. 7!) are now in progress in this laboratory.

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SURFACE MOLECULAR STRUCTURE FROM ION-NEUTRALIZATION SPECTROSCOPY OF ELECTRONS IN SURFACE ORBITALS

H. D. Hagstrum and G. E. Becker
Bell Telephone Laboratories, Murray Hill, New Jersey 07974

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It is shown that the ion-neutralization spectroscopy determines the orbital energy spectra of surface molecules formed of adsorbed foreign atoms and surface atoms of the substrate. For free molecules such a spectrum yields information concerning molecular symmetry and structure as well as ionicity within the molecule. Using also measured work-function change in adsorption, specific conclusions are drawn concerning O, S, and Se adsorbed in $p(2\times2)$ and $c(2\times2)$ arrangements on Ni (100).

In surface physics and chemistry we have not to this point possessed sufficiently powerful experimental methods to determine the local symmetry and molecular structure in which foreign atoms are adsorbed on solids. In its present state of development, low-energy electron diffraction determines only the symmetry of the pattern in which the local structure or surface molecule repeats itself over the surface. Work-function change on adsorption yields an important parameter but its value depends both on ionicity and structure. In this Letter we demonstrate that the ion-neutralization spectroscopy[1] determines the orbital energy spectrum of electrons in surface bonds whose form and energy position, for certain systems at least, enable us to draw specific conclusions about the structure and symmetry of the local molecular arrangement. We present experimental results and conclusions for O, S, and Se adsorbed in $p(2\times2)$ and $c(2\times2)$ arrangements on Ni (100).

The orbital energy spectrum is determined by means of the ion-neutralization spectroscopy discussed extensively elsewhere.[1] The method determines a function $U(\zeta)$, called the transition density, which includes both state density and transition probability factors. ζ is the energy measured positively downward into the filled band from zero at the Fermi level. When a foreign atom is adsorbed on a surface, the transition probability of the Auger process is enhanced at the energies of the orbitals of electrons in the surface bonds. The orbital energy states which lie in the energy range of the allowed states of the solid become broadened resonances or virtual states. States lying outside (below) the allowed range in the solid are broadened by the effect of molecular vibration on orbital energy and by lifetime and other effects having to do with the ion-neutralization process. We expect such states to be sharper than the resonances or virtual states.

In Fig. 1 are shown the transition-density functions obtained for clean Ni (100), curve 1, and for this surface with $p(2\times2)$ and $c(2\times2)$ surface arrangements involving $X \equiv$ O, S, or Se, curves 2, 2′ to 4, 4′. We note first that the Ni d band, to be seen just below the Fermi level ($\zeta = 0$) in curve 1, is drastically reduced in intensity in curves 2, 3, and 4, which correspond to a half-monolayer of the adsorbate X, and somewhat less so in curves 2′, 3′, and 4′ which correspond to a quarter-monolayer of X. This reduction in d-band intensity is direct experimental evidence that the ion-neutralization process is sensitive to wave-function magnitude in the surface layer of the solid primarily. The $U(\zeta)$ functions in Fig. 1 are shown by a dashed line in the energy range $\zeta > 9$ eV because for several good reasons the data are not accurately reproducible here. However, the presence of a peak in this region in curves 3 and 4 and its absence in curves 3′ and 4′ is a reproducible experimental conclusion.

The structure which appears in curves 2, 2′ to 4, 4′ of Fig. 1, not including the residual Ni d-band peak, is taken to be the molecular orbital energy spectrum of the electrons in the bonds of what may be called surface molecules formed of the adsorbate atom and atoms of the first layer