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INTERLAYER EXCHANGE COUPLING AND DAMPING PROCESSES IN COUPLED TRILAYER SYSTEMS

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Abstract: In ferromagnetic resonance the most reliable determination of the interlayer exchange coupling is only possible if a full angular dependence of the resonance field can be measured and explained by correct angle-resolved resonance equations. We show the calculation procedure and provide an example of the angular FMR dependence and IEC oscillations with spacer thickness for Ni/Cu/Ni(Co) trilayers. The second point we would like to address is the effect of the spin pumping phenomena on the damping processes. We show that the resonance linewidth ΔH follows the behavior of the IEC, *i.e.*, it oscillates with the spacer thickness. Besides, in our X-ray magnetic circular dichroism studies we have found that the Curie temperature can be shifted due to the IEC. The importance of this effect on the FMR linewidth is shown, too.

1. INTRODUCTION

Since the discovery of the interlayer exchange coupling (IEC) in a trilayer system composed of two ferromagnetic (FM) films separated with a nonmagnetic spacer by Grünberg *et al*. [1] in 1986 the coupling has been observed qualitatively and quantitatively employing a variety of experimental techniques. Most of the methods provide either only the sign of the coupling parameter or concern the unsaturated state characterized by the presence of magnetic domains. A method free of most of the drawbacks is ferromagnetic resonance (FMR). It can provide the value of the coupling strength in absolute units. It results from the fact that the precession of the two degenerate uniform modes (assuming two identical uncoupled FM films) splits into two different resonance excitations after switching on the interlayer interaction. The main mode is created by the in-phase precession of the two magnetizations (acoustical mode) and the secondary mode corresponds to the out-of-phase precession (optical mode). The magnetic field separation of the two modes is a direct measure of the coupling strength [2-4]. However, a proper extraction of the coupling constant $J_{\rm inter}$ requires the use of adequate resonance equations including all the parameters characterizing the system, *i.e*., the magnetocrystalline and shape anisotropy, the magnetizations, *g*-factors and the interlayer exchange parameter. Usually a simplified case is investigated by performing the FMR experiment with the external magnetic field either parallel or normal to the film plane. A more reliable value of the IEC can be extracted from full angular measurements for both the acoustical and the optical mode. However, this requires the determination of the equilibrium

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angles for each orientation of the magnetic field [3, 4]. In this paper we describe one of the possible approaches for searching the solutions for the acoustical and the optical excitations. An example for the Ni/Cu/(Ni)Co films is also shown. The experimental measurements of the ferromagnetic resonance are carried out fully *in situ*, i.e., the first FM film can be characterized by FMR including the angular dependence and subsequently the next parts of the sample can be evaporated and studied by FMR without breaking the vacuum.

The above considerations concern only the static properties of the ferromagnetic resonance. However, from the point of view of practical applications, *e.g.* in spintronics, also the dynamic is important. In the case of FMR the information about the relaxation processes is hidden in the linewidth of the resonance signal. Various intrinsic and extrinsic contributions are well recognized for a single uncoupled FM film but there are still some challenges in the case of coupled trilayers. In the second part of the paper we address the problem of the additional broadening of the FMR linewidth ΔH due to the spin current pumped by the precessing magnetizations [5-7]. The sample preparation is described elsewhere [8].

2. ANGULAR DEPENDENCE OF FMR IN COUPLED TRILAYERS

The free energy describing a trilayer system composed of two ferromagnetic films separated with a nonmagnetic spacer takes the form:

$$\begin{aligned} F = \sum_{i=1}^{2}\Big[& -d_i M_i H\left(\sin\theta_i \sin\theta_H \cos(\varphi_i-\varphi_H)+\cos\theta_i\cos\theta_H\right) + \\ & -d_i\left(2\pi M_i^2-K_2^i\right)\sin^2\theta_i-\frac{1}{2}d_i K_{4\perp}^i\cos^4\theta_i + \\ & -\frac{1}{8}d_i K_{4||}^i\left(3+\cos 4\varphi_i\right)\sin^4\theta_i-d_i K_{2||}^i\sin^2\theta_i\cos^2\left(\varphi_i-\delta_i\right)\Big] + \\ & -J_{\mathrm{inter}}[\sin\theta_1\sin\theta_2\cos\left(\varphi_1-\varphi_2\right)+\cos\theta_1\cos\theta_2] + \\ & -J_{\mathrm{biq}}[\sin\theta_1\sin\theta_2\cos\left(\varphi_1-\varphi_2\right)+\cos\theta_1\cos\theta_2]^2 \end{aligned} \tag{1}$$

where $i = 1, 2$. d_i is the thickness and M_i the magnetization of the FM films. K_2^i represents the perpendicular anisotropy and in the case of the cubic anisotropy, K_4, the tetragonal distortion is taken into consideration, *i.e.*, K_4 is separated into the in-plane $K_{4||}^i$ and the out-of-plane $K_{4\perp}^i$ component. For the sake of generality we also include the appropriate terms for the in-plane uniaxial anisotropy leaving the possibility of a shift by angle δ_i between the easy axis of the cubic and the uniaxial contribution. As usually θ_i, φ_i and θ_H, φ_H are the polar and azimuthal angles of the magnetizations and magnetic field, respectively. Apart from the bilinear term J_{inter} also the biquadratic coupling J_{biq} is included in Eq. (1). However, it is negligible in the trilayer systems discussed in this paper because this term is usually related to fluctuations of the spacer thickness [9] and our epitaxial films are of high quality. Besides, a small biquadratic contribution can hardly be distinguishable from a small modification of the present anisotropy terms [10]. The equation of motion in the free energy formalism for a system of two FM films in a matrix form composed of second derivatives of the free energy can be found in Ref. 2. After evaluating these second derivatives the following resonance formula is obtained [11]:

$$a\omega^4 + b\omega^2 + c = 0 \tag{2}$$

where

$$a = \frac{d_1^2 M_1^2 \sin^2\theta_1}{\gamma_1^2}\,\frac{d_2^2 M_2^2 \sin^2\theta_2}{\gamma_2^2} \tag{3}$$

$$\begin{aligned} b =& \left(F_{\theta_1\varphi_1}^2 - F_{\theta_1\theta_1}F_{\varphi_1\varphi_1} \right) \frac{d_2^2 M_2^2 \sin^2\theta_2}{\gamma_2^2} + \left(F_{\theta_2\varphi_2}^2 - F_{\theta_2\theta_2}F_{\varphi_2\varphi_2} \right) \frac{d_1^2 M_1^2 \sin^2\theta_1}{\gamma_1^2} + \\ &+ 2\left(F_{\theta_1\varphi_2}F_{\theta_2\varphi_1} - F_{\theta_1\theta_2}F_{\varphi_1\varphi_2} \right) \frac{d_1 d_2 M_1 M_2 \sin\theta_1 \sin\theta_2}{\gamma_1\gamma_2} \end{aligned} \tag{4}$$

$$\begin{aligned} c =\ & F_{\theta_1\theta_2}^2 F_{\varphi_1\varphi_2}^2 + F_{\theta_1\varphi_1}^2 F_{\theta_2\varphi_2}^2 + F_{\theta_1\varphi_2}^2 F_{\theta_2\varphi_1}^2 - F_{\theta_1\theta_2}^2 F_{\varphi_1\varphi_1} F_{\varphi_2\varphi_2} - F_{\varphi_1\varphi_2}^2 F_{\theta_1\theta_1} F_{\theta_2\theta_2} + \\ & - F_{\theta_2\varphi_2}^2 F_{\theta_1\theta_1} F_{\varphi_1\varphi_1} - F_{\theta_1\varphi_2}^2 F_{\theta_2\theta_2} F_{\varphi_1\varphi_1} - F_{\theta_1\varphi_1}^2 F_{\theta_2\theta_2} F_{\varphi_2\varphi_2} - F_{\theta_2\varphi_1}^2 F_{\theta_1\theta_1} F_{\varphi_2\varphi_2} + \\ & + F_{\theta_1\theta_1} F_{\varphi_1\varphi_1} F_{\theta_2\theta_2} F_{\varphi_2\varphi_2} + 2F_{\theta_1\theta_1} F_{\varphi_1\varphi_2} F_{\theta_2\varphi_1} F_{\theta_2\varphi_2} + 2F_{\theta_1\varphi_1} F_{\theta_1\varphi_2} F_{\varphi_1\varphi_2} F_{\theta_2\theta_2} + \\ & + 2F_{\theta_1\theta_2} F_{\theta_1\varphi_2} F_{\varphi_1\varphi_1} F_{\theta_2\varphi_2} + 2F_{\theta_1\theta_2} F_{\theta_1\varphi_1} F_{\theta_2\varphi_1} F_{\varphi_2\varphi_2} - 2F_{\theta_1\theta_2} F_{\varphi_1\varphi_2} F_{\theta_1\varphi_2} F_{\theta_2\varphi_1} + \\ & - 2F_{\theta_1\varphi_1} F_{\theta_2\varphi_2} \left(F_{\theta_1\theta_2} F_{\varphi_1\varphi_2} + F_{\theta_1\varphi_2} F_{\theta_2\varphi_1} \right) \end{aligned} \tag{5}$$

In these formulas $\omega = 2\pi f$ is the frequency and γ_i represents the gyromagnetic ratio.

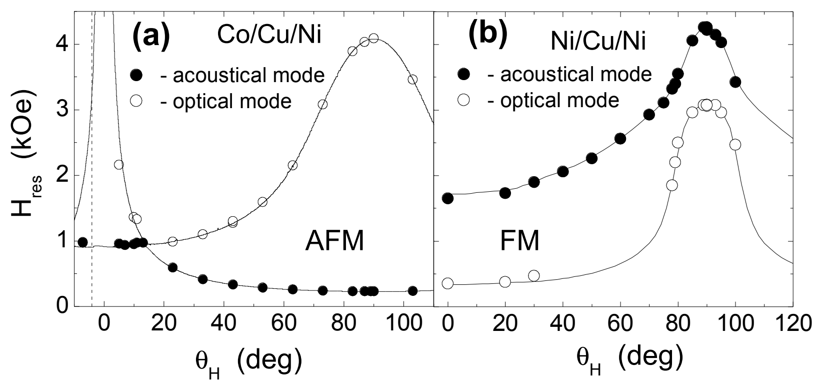


Fig. 1. Measured and calculated angular dependence of the acoustical and optical mode for (a) weakly AFM coupled $Ni_9Cu_{25}Co_2$ (b) FM coupled $Ni_8Cu_5Ni_9$ trilayers (subscripts denote the thickness in monolayers – ML)

In the usual experimental setup with H in the easy in-plane direction one may set $\theta_i = \theta_H = 90°$ and $\varphi_i = \varphi_H = 0°$, which leads to much simplified Eqs. (3-5). We intend to calculate the full angular dependence, therefore for each θ_H the equilibrium angles θ_i have to be calculated from the minimum conditions $F_{\theta_i\theta_i} > 0$ and $F_{\theta_1\theta_1}F_{\theta_2\theta_2} - F^2_{\theta_1\theta_2} > 0$. We assume that due to the small cubic in-plane anisotropy $\varphi_i = \varphi_H = 0$ can be applied, *i.e.* the magnetizations are constrained within a vertical plane. The equilibrium angles are determined by the steepest-descent method (gradient analysis) like in the case of demagnetization process calculations [12]. For each value of H (starting from saturation) and θ_i the resonance equation is controlled and all the possible resonance modes between H_{max} and $H = 0$ are localized. Figure 1(a) shows an example of the angular dependence for the $Ni_9Cu_{25}Co_2$ trilayer and Fig. 1(b) corresponds to the case of $Ni_8Cu_5Ni_9$ [11] (subscripts denote the thickness in monolayers – ML). For the former there is a weak antiferromagnetic coupling and for the latter it is ferromagnetic. The solid lines represent a fit using the above outlined calculations. For Fig. 1(a) the split of the dependences stems from the coupling creating the acoustical and the optical mode. In the absence of coupling the Ni and Co angular dependencies would cross each other due to the different anisotropies with the out-of-plane direction being an easy axis for Ni and hard axis for Co [4, 8].

4. ENHANCEMENT OF MAGNETIZATION DAMPING BY THE SPIN-PUMPING EFFECT

Till now we have shown results and analysis for the static properties of FMR. In this section the dynamics is investigated, which is also influenced by the interlayer interaction.

The basic contributions to the FMR linewidth can be written as:

$$\Delta H(\omega,\alpha,x_i) = \Delta H^D + \left|\frac{\partial H(\omega,x_i)}{\partial x_i}\right|\Delta x_i + \frac{2}{\sqrt{3}}\frac{G}{\gamma^2 M}\frac{\omega}{\cos(\beta-\beta_H)} . \tag{6}$$

The first right-hand term is due to the lattice defects and the second term describes the spread of any material parameters x_i. In some cases these inhomogeneities can lead to the broadening of ΔH by two magnon scattering [13, 14]. Also impurity spins at the interface can be a source of a slow relaxation mechanism [15]. The third contribution is intrinsic in origin and represents damping by a viscosity-like mechanism described by the Gilbert parameter G [16]. M is the magnetization, γ represents the gyromagnetic ratio and β and β_H are the magnetization and magnetic field angles in respect to the easy magnetization axis within a studied plane.

However, there are additional contributions possible if a ferromagnetic film is in contact with a nonmagnetic one and a transfer of torque occurs (so called spin pumping). The resulting spin current is

$$\mathbf{I}_S^{\text{pump}} = \frac{\hbar}{4\pi}\left(A_r \mathbf{m}\times\frac{d\mathbf{m}}{dt} - A_i\frac{d\mathbf{m}}{dt}\right) \tag{7}$$

with A_r and A_i being the real and the imaginary conductivities, respectively [5-7]. It is at once visible that the first term is exactly of the Gilbert type (the second term gives only a negligible shift of the resonance field), therefore the general equation of motion can be written as

$$\frac{d\mathbf{m}}{dt} = -\gamma \mathbf{m} \times \mathbf{H}_{\text{eff}} + (\alpha + \alpha')\, \mathbf{m} \times \frac{d\mathbf{m}}{dt} \tag{8}$$

which means that the spin current enhances the damping parameter $\alpha = G/\gamma M$. This additional Gilbert-like broadening has been verified for the Fe/Au/Fe/GaAs trilayers by Heinrich *et al.* [7] and recently in our studies for the Ni/Cu/Ni(Co) trilayers [17]. There are several conditions determining the contribution of the spin current: (i) The thickness of the FM film has to be larger than the transverse spin-coherence length $\lambda_{SC} = \pi/(k_\uparrow - k_\downarrow)$, where $k_\uparrow$, $k_\downarrow$ denote the Fermi wave vectors. (ii) If the transmission t_{NM} from the non-magnet to the ferromagnet NM=>FM is small (t_{NM}=>0) a perfect spin sink is provided, *i.e.* the back current created by the spin accumulation in the NM film is small ($\mathbf{I}_S^{back} = 0$). Then the NM thickness d_{NM} is not important and the increased Gilbert damping is observed. (iii) If the transmission is large, then for $d_{NM} \ll \lambda_{SF}$ (λ_{SF} is a spin-flip length within the NM) the spin accumulation leads to $\mathbf{I}_S^{back} > 0$ giving the compensation $\mathbf{I}_S^{pump} - \mathbf{I}_S^{back}$. To observe the increased Gilbert damping d_{NM} has to be increased much above λ_{SF}. (iv) Even for $d_{NM} \ll \lambda_{SF}$ the presence of the additional damping can be verified by its sudden disappearing at a compensation point for equal resonance conditions when H_{res}(FM1) = H_{res}(FM2). The last possibility is illustrated schematically in Fig. 2(a) and can occur when, *e.g.*, the angular dependences of the two FM films cross or approach each other [7, 17]. An example of such an angular dependence is provided in Fig. 2(b) for a large spacer thickness (zero interlayer coupling).

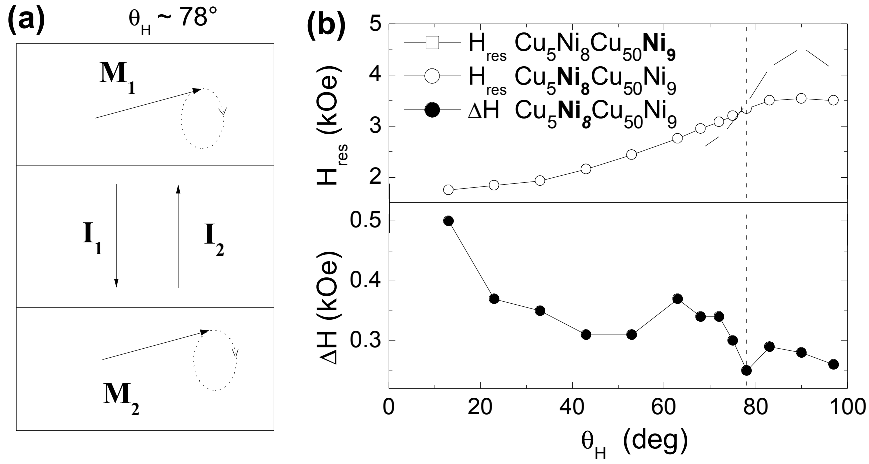
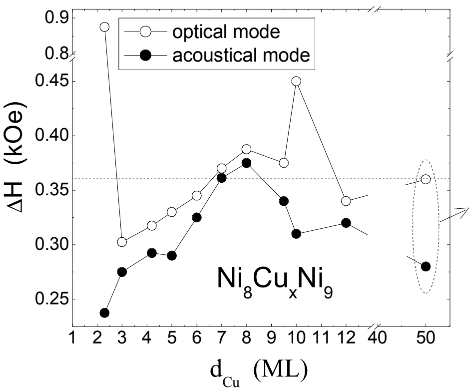


Fig. 2. (a) A sketch of the simultaneous precession of magnetizations at the crossing of the angular dependences of the resonance fields. (b) The angular dependence of the resonance field and the linewidth for the uncoupled Ni/Cu/Ni trilayer. Lines are a guide to the eye. The angular dependence concerns always the film marked out in bold font. The drop of linewidth is visible at 78º

However, the pumped spin-current is equivalent to the dynamical part of the interlayer coupling; therefore, one can expect that this contribution to the damping should likewise IEC exhibit an oscillatory behavior. We have observed this effect and it is presented in Fig. 3 and Fig. 4 for the linewidth of the acoustical and the optical mode. In the case of these modes the two magnetizations precess together in-phase or out of phase ‘pumping’ the spin currents simultaneously. Assuming that the standard Gilbert contribution is equal for both Ni films (of nearly equal thickness) the difference in the linewidth for the two modes is:

$$\Delta H^{ac} - \Delta H^{opt} = \left[\Delta H^{Ni}_{Gilb} + \Delta H^{ac}_{SP}\right] - \left[\Delta H^{Ni}_{Gilb} + \Delta H^{opt}_{SP}\right] = \Delta H^{ac}_{SP} - \Delta H^{opt}_{SP} \tag{9}$$



ΔH(Ni$_9$) (kOe)	preparation step
0.31	$Cu_{10}Ni_9$/Cu(001)
0.32	$Cu_{50}Ni_9$/Cu(001)
0.36 0.28(Ni$_8$)	$Cu_5Ni_8Cu_{50}Ni_9$/Cu(001)

Fig. 3. Linewidth of the optical and the acoustical mode versus the spacer thickness. On the right hand side the evolution of the linewidth of the Ni_9 film is shown for various steps of the trilayer preparation (uncoupled system, spacer of 50 ML). The details are explained in the text

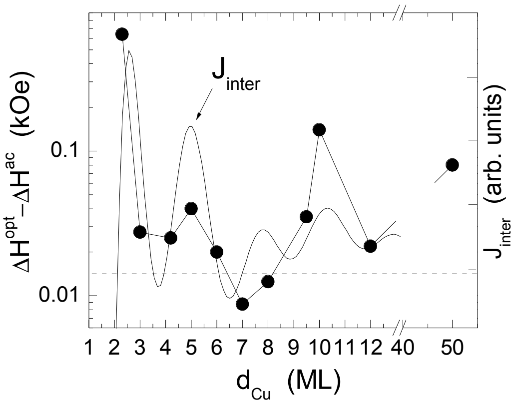


Fig. 4. Oscillatory behavior of the difference between the linewidth of the acoustical and the optical mode representing mainly the contribution due to the pumped spin-currents. The oscillations of IEC calculated from Bruno model and reflecting the measured IEC oscillations [11] are displayed for comparison. The vertical scale of J_{inter} is in arbitrary units and linear, therefore only the periods can be compared

Hence, the difference in the linewidth for both modes is a good quantity to test the oscillatory behavior of ΔH. Due to the larger current compensation for the acoustical mode the oscillatory behavior is mainly provided by the optical part, $\Delta H^{\mathrm{opt}}_{SP}$. This difference is plotted in Fig. 4 and shows a period of oscillations roughly consistent with our previous IEC studies (the solid line indicated with J_{inter} shows the IEC oscillations calculated basing on the Bruno model [11]). Obviously, the amplitude attenuation can be quite different for the linewidth than for the IEC.

The ΔH oscillations are only roughly visible because the difference in Eq. (9) is not the only possible contribution (see below) and besides, the determination of the linewidth is not as precise as in the case of the line position employed for IEC extracting.

The asymmetry of the spin currents is mainly caused by the presence of the second FM film working as the spin sink. This effect is illustrated on the right hand side of Fig. 3 for the limit of a thick spacer (50 ML). This implies a zero coupling, therefore the experimental points can be ascribed at this thickness to the Ni_8 (full circle) and Ni_9 (open circle) instead of the acoustical and optical mode. Taking advantage of our fully *in situ* preparation and step by step measurements we can see that the increase in the Cu spacer thickness from 10 ML to 50 ML gives only a small decrease in $\mathbf{I}_S^{\text{back}}$ (increase of ΔH from 0.31 to 0.32 kOe). Already the deposition of the second Ni film leads to the more significant broadening of ΔH (from 0.31 to 0.36 kOe), *i.e.* a larger net spin current is present. The $\Delta H(d_{Cu})$ dependence in Fig. 3 can be separated into two regions. Above about 40 ML the linewidth of each mode is represented by the Gilbert term and the non-oscillating contribution due to the spin-pumping effect (other terms of Eq (6) can be neglected or are constant because only the in-plane ΔH is considered). For thinner spacers ΔH_{SP} is oscillating as discussed above. In general, for the thinnest spacers additional contributions due to the coupling inhomogeneity and/or due to the T_C shift can be important. The latter is described below and both are negligible for the systems studied.

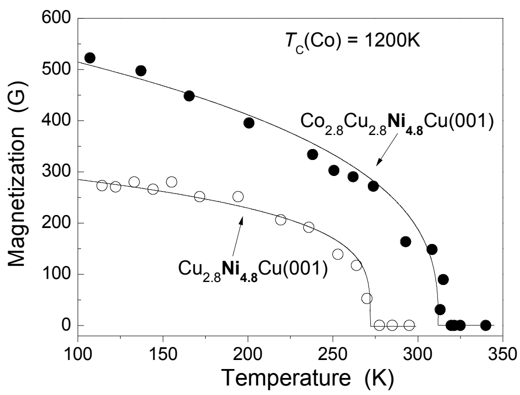


Fig. 5. XMCD studies of the T_C shift of the Ni film due to the deposition of the second FM film (Co). This shift is caused by the strong IEC

In general, a temperature dependence of ΔH for a single FM film is changing with thickness because of the change in T_C (dimensional effect). Recently, we have shown that the IEC returns partly the third dimension, *i.e.*, due to the IEC a trilayer system behaves as a thicker film of higher T_C. The T_C-shift after deposition of a second FM film (Co) is shown in Fig. 5 basing on XMCD measurements [18]. We have shown experimentally [19], and it was also predicted theoretically [20] that this shift exhibits an oscillatory behavior. Therefore, this modification of T_C in connection with the above mentioned temperature dependence of ΔH implies a change of ΔH, which is an additional oscillatory contribution to the linewidth, which we expected for ultrathin FM films (thinner than in the current studies) and strong interlayer coupling values.

5. CONCLUSIONS

We have shown a procedure for an effective calculation of the angular dependence of the acoustical and the optical mode measured in the ferromagnetic resonance experiment on coupled trilayers. Such angular studies enable the precise extraction of the coupling strength and determination of its oscillations as a function of the spacer thickness. A spin current pumped by the precessing magnetizations leads to a broadening of the linewidth. This additional damping exhibits an oscillatory behavior like the interlayer coupling. It has been predicted that the T_C-shift due to the IEC can also be important for the linewidth value of ultrathin ferromagnetic films.

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