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Enhancement of the magneto-optical Kerr angle in nonlinear optical response

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Using an electronic theory we present calculations of the magneto-optical Kerr rotation *angle* in second-harmonic generation (SHG). For the longitudinal and polar Kerr configuration and for arbitrary angles of incidence it is found that the Kerr angle in SHG may be enhanced by up to one order of magnitude compared to the linear Kerr angle. This enhancement is caused by interband and intraband transitions (plasmons), which in the linear case suppress the Kerr rotation in the optical range. Our results will be useful for a microscopic study of two-dimensional magnetism.

Nonlinear optics is of general interest and may in particular become an important tool for studying fundamental problems of magnetism at surfaces, interfaces, and in films. Since second-harmonic generation (SHG) requires inversion-symmetry breaking, it is generally far more surface sensitive in inversion-symmetric media than linear optical studies and thus particularly suited to determine magnetic moments, magnetic ordering, magnetic easy axes, and spin-orbit coupling. In multilayers, SHG benefits from both the lack of inversion symmetry at each interface generating a frequency-doubled signal and the large penetration depth of the fundamental light giving access to buried interfaces. Presently, SHG is, for example, the only probe of magnetism at the interface of two magnetic films. Since there is no theory available so far, it is the goal of this paper to show using an electronic theory how magnetism affects the incoming light polarization and to determine the frequency-dependent Kerr rotation and dichroism. For this we have also to extend the Fresnel formulas to the nonlinear magnetic case, which seems of general interest by itself. Since in the linear case the Kerr rotation is suppressed by bulk interband and intraband transitions (plasmons), one expects in the nonlinear case with only surface optical response an enhancement of the Kerr rotation. Indeed we find a large enhancement of the Kerr rotation of the light polarization as a general phenomenon in nonlinear magneto-optics. This might explain then also various recent experimental results on garnets and Heusler alloys.[1–3] Since, also with regard to the high potential for technological application,[4,5] the search for large Kerr rotations in linear optics has been a longstanding subject of intense theoretical and experimental investigations, such an enhancement of the Kerr rotation in the nonlinear magneto-optical Kerr effect (NMOKE) possibly opens a new route for applications involving readily available materials, such as iron, without resorting to low temperatures, large magnetic fields, or binary and ternary magnetic alloys.[6] Thus, our theory may become the basis for a unified use of SHG as a tool for a comprehensive study of surface and interface magnetism including magnetic anisotropy, spin-orbit coupling, etc.

Using electrodynamical theory the linear and nonlinear polarizations are expressed by the susceptibilities. The influence of magnetism on SHG is shown best by determining the change of the polarization of the incoming light. For this we use the wave equation (j=1,2)

$$\nabla \times \nabla \times \mathbf{E}^{(j)}(j\omega) + \frac{\varepsilon(j\omega)}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{E}^{(j)}(j\omega) = -\frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}\mathbf{P}^{(2)}\,\delta_{2j}\ , \quad (1)$$

with the nonlinear polarization

$$\mathbf{P}^{(2)}(2\omega) = \chi^{(2)}(2\omega) : \mathbf{E}^{(1)}(\omega)\cdot\mathbf{E}^{(1)}(\omega) \quad (2)$$

as a source term. To link SHG to the spin-polarized electronic structure and to understand the microscopic origin of the Kerr rotation, we use an electronic theory to determine $\chi^{(2)}(\omega)$. Note, Eq. (1) is in the linear case without the source term a homogeneous wave equation leading to left- and right-handed circularly polarized waves as eigenmodes with complex refraction indices N_+ and N_- as eigenvalues controlling the optical response. In the nonlinear case the source term in Eq. (1) yields an inhomogeneous wave equation having solutions that no longer relate to N_+ and N_- but directly to the surface response function $\chi^{(2)}$, which does not correspond to indices of refraction.

We use now for the calculation of the Kerr rotation the law of reflection and decompose $\mathbf{E}^{(j)}(j\omega)$ into left- and right-handed circularly polarized light $\mathbf{E}^{(\pm)(j)}(j\omega)$ due to the magnetic birefringence. One obtains for the complex Kerr angle with real part $\phi_K^{(j)}$ and ellipticity $\varepsilon_K^{(j)}$ (amplitude of field $\mathbf{E}_r^{(\pm)}$: $\mathbf{E}_{r,a}^{\pm}$)

$$\tan\psi_K^{(j)} = \phi_K^{(j)} + i\varepsilon_K^{(j)} = i\frac{\mathbf{E}_{r,a}^{(+)(j)}(j\omega) - \mathbf{E}_{r,a}^{(-)(j)}(j\omega)}{\mathbf{E}_{r,a}^{(+)(j)}(j\omega) + \mathbf{E}_{r,a}^{(-)(j)}(j\omega)}. \quad (3)$$

Further evaluation of Eq. (3) requires the specification of the Kerr configuration. We choose the *longitudinal* Kerr configuration as shown in inset (a) of Fig. 2 for arbitrary angles of incidence. Note, other configurations may be analyzed similarly. In this configuration the magnetization vector **M** lies parallel to the optical plane and parallel to the sample surface. We assume p-polarized incident light. For **M**=**0**, $\mathbf{P}^{(2)}$ perpendicular to the sur-

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face gives the maximum SHG yield.[7] Inset (b) in Fig. 2 shows the definition of the linear and nonlinear Kerr angles with respect to the reflected beam of frequency ω and 2 ω, respectively.

To express the field amplitude $\mathbf{E}_{r,a}^{(\pm)(j)}(j\omega)$ of the reflected beam by the incident field amplitude $\mathbf{E}_{i,a}^{(\pm)(j)}(j\omega)$ we use the usual Fresnel formulas[8] for the linear case and derive such expressions for the nonlinear case extending results by Bloembergen and Pershan[9] to the case of magnetic surfaces. We find for the nonlinear magnetic Fresnel formula in p-polarization the result[10]

$$\begin{aligned} E_r^{(2)\pm} =& -\frac{\chi^{(2)\pm}[E_i^{(1)}\sin^2\Theta_i]^2\sin\Theta_i}{\varepsilon_0 c^2}\,\frac{1}{[1+\chi^{(1)\pm}(2\omega)]S_1^{\pm}(\Theta_i)+[1+\chi^{(1)\pm}(\omega)]S_2^{\pm}(\Theta_i)}\\ &\times\frac{1}{[1+\chi^{(1)\pm}(2\omega)]\cos\Theta_i+S_2^{\pm}(\Theta_i)}\left\{\sin^2\Theta_i+\frac{1+\chi^{(1)\pm}(2\omega)}{1+\chi_0^{(1)}(2\omega)}S_1^{\pm}(\Theta_i)S_2^{\pm}(\Theta_i)\right.\\ &\left.+\frac{\chi^{(1)\pm}(2\omega)-\chi_0^{(1)}(2\omega)}{\chi^{(1)\pm}(\omega)-\chi_0^{(1)}(2\omega)}\left[\frac{1+\chi^{(1)\pm}(2\omega)}{1+\chi_0^{(1)}(2\omega)}[1+\chi^{(1)\pm}(\omega)]-2\sin^2(\Theta_i)\right]\;S_1^{\pm}(\Theta_i)S_2^{\pm}(\Theta_i)\right\}\\ \equiv& -\frac{P^{(2)\pm}\sin\Theta_i}{\varepsilon_0 c^2}\,\frac{1}{F_3^{\pm}F_2^{\pm}}\,\{F_1^{\pm}\}\,, \end{aligned} \tag{4}$$

with $S_j^{\pm}(\Theta_i)\equiv\sqrt{1+\chi^{(1)\pm}(j\omega)-\sin^2\Theta_i}$ for j=1,2. The $F_n^{\pm}$ follow by comparison (n=1,2,3). Here the tensors $\chi^{(1)\pm}=\chi_0^{(1)}\pm i\chi_1^{(1)}\sin\Theta_i$ and $\chi^{(2)\pm}=\chi_0^{(2)}\pm\chi_1^{(2)}$ are decomposed in diagonal ("nonmagnetic") and off-diagonal ("magnetic") contributions. Due to the source term in Eq. (1) the i is missing in $\chi^{(2)\pm}$. We find then for the longitudinal geometry the linear Kerr angle

$$\begin{aligned} \tan\psi_K^{(1)}(\omega) =& -\frac{\chi_1^{(1)}(\omega)}{\chi_0^{(1)}(\omega)}\frac{\sin\Theta_i\cos\Theta_i}{\sqrt{\cos^2\Theta_i+\chi_0^{(1)}(\omega)}}\\ &\times\frac{\cos(2\Theta_i)+\chi_0^{(1)}(\omega)}{\cos(2\Theta_i)+\chi_0^{(1)}(\omega)\cos^2\Theta_i}\,, \end{aligned} \tag{5}$$

and for the nonlinear Kerr angle

$$\tan\psi_K^{(2)}(\omega) = i\frac{\chi^{(2)+}F_1^+F_2^-F_3^- - \chi^{(2)-}F_1^-F_2^+F_3^+}{\chi^{(2)+}F_1^+F_2^-F_3^- + \chi^{(2)-}F_1^-F_2^+F_3^+}\,. \tag{6}$$

Eqs. (5) and (6) are the basis for determining the ω and Θ_i dependence of the linear and nonlinear Kerr rotation. Since $\chi_1^{(j)}\ll\chi_0^{(j)}$, Eq.(6) may be expanded in powers of $(\chi_1^{(j)}/\chi_0^{(j)})$ and linearized. One gets using $F_k^{\pm}=F_{k0}\pm F_{k1}$ (k=1,2,3)

$$\tan\psi_K^{(2)} = i\left(\frac{\chi_1^{(2)}}{\chi_0^{(2)}}+\frac{F_{11}}{F_{10}}-\frac{F_{21}}{F_{20}}-\frac{F_{31}}{F_{30}}\right)\,, \tag{7}$$

with $F_{k0}=F_k^{\pm}(F_{k1}=0)$, $(k=1,2,3)$,

$$\begin{aligned} F_{11} = i\sin\Theta_i\Bigg[&\chi_1^{(1)}(2\omega)S_{10}(\Theta_i)S_{20}(\Theta_i)\\ &+\chi_1^{(1)}(2\omega)\frac{S_{10}(\Theta_i)}{2S_{20}(\Theta_i)}+\chi_1^{(1)}(\omega)\frac{S_{20}(\Theta_i)}{2S_{10}(\Theta_i)}\\ &+\frac{\chi_1^{(1)}(2\omega)[1+\chi_0^{(1)}(\omega)-2\sin^2(\Theta_i)]}{\chi_0^{(1)}(\omega)-\chi_0^{(1)}(2\omega)}\Bigg]\,, \end{aligned}$$

$$F_{21} = i\sin\Theta_i\left[\chi_1^{(1)}(2\omega)\left(\cos\Theta_i+\frac{1}{2S_{20}(\Theta_i)}\right)\right]\,,$$

$$\begin{aligned} F_{31} = i\sin\Theta_i\Bigg[&\chi_1^{(1)}(2\omega)\left(S_{10}(\Theta_i)+\frac{1+\chi_0^{(1)}(\omega)}{2S_{20}(\Theta_i)}\right)\\ &+\chi_1^{(1)}(\omega)\left(S_{20}(\Theta_i)+\frac{1+\chi_0^{(1)}(2\omega)}{2S_{10}(\Theta_i)}\right)\Bigg]\,, \end{aligned} \tag{8}$$

where $S_{j0}(\Theta_i)\equiv\sqrt{1+\chi_0^{(1)}(j\omega)-\sin^2(\Theta_i)}$.

Note, the first term in Eq. (7) results from $\chi^{(2)+}\neq\chi^{(2)-}$, and has been previously neglected.[1] This term, however, gives the main contribution to the nonlinear Kerr rotation. It vanishes in the case of inversion symmetry in the bulk material, but not at the surface. The F_{k1} and F_{k0}, however, depend only on *linear* susceptibilities. Hence, nonlinearity exhibits a much stronger surface sensitivity which is responsible for making the NMOKE a very useful tool for studying surface magnetism. It is interesting, that for all configurations the factor $1/\sqrt{\cos^2\Theta_i+\chi_0^{(1)}(\omega)}$, which causes the small $\psi_K^{(1)}$ in Eq. (5), is not present in the nonlinear case [Eq. (7)]. Despite the complex dependence of the parameters it is clear that the nonlinear Kerr rotation is for all Θ_i always enhanced by a factor $\sqrt{\cos^2\Theta_i+\chi_0^{(1)}(\omega)}$. Note, this enhancement can be traced back to the source term of the wave equation (1), where $P^{(2)}\sim\chi^{(2)}$ and $\chi^{(2)}$ depends sensitively on the magnetic properties at the surface. This is the mathematical manifestation of the different character of the solution of the homogeneous and inhomogeneous wave equations Eq. (1) as pointed out already and of the different physics involved in linear and nonlinear optics. In the nonlinear case only the surface contributes to the optical response. Thus, the destructive contributions to the Kerr rotation by bulk inter- and intraband transitions are avoided. Furthermore, it follows clearly from Eq. (7) that the interaction between the light and magnetism is mediated by the spin-orbit coupling. For vanishing spin-orbit coupling constant $\lambda_{\mathrm{s.o.}}$ one gets $\chi_1^{(2)}=F_{k1}=0$ (k=1,2,3) and thus $\psi_K^{(2)}=0$. All F_{k0} and $\chi_0^{(j)}$ are independent of spin-orbit coupling.

RAPID COMMUNICATIONS

To demonstrate now numerically the effect of magnetism on SHG, in particular the frequency-dependent enhancement of the Kerr rotation, we use Eqs. (5) and (6) assuming $\Theta_i = 45°$. The functions $F_k^{\pm}$ (k=1,2,3), see Eq. (4), are determined by the susceptibilities $\chi_i^{(j)}$ (i=0,1; j=1,2). These have been calculated previously by us for Fe.[11] The obtained results should be representative for transition metals. Only for the nonmagnetic linear susceptibility $\chi_0^{(1)}$ we include additively interband and intraband contributions. For the intraband contribution, we assume as usual a non-spin-split conventional Drude form $\chi_{0,\text{intra}}^{(1)} = \omega_0^2/[\omega(\omega + i/\tau)]$, using τ=9.12×10^{-15} sec and $\hbar\omega_0$=0.74 eV. We neglect Drude contributions to the magnetic susceptibilities since the d electrons dominate the electronic spin polarization. For the nonlinear nonmagnetic susceptibility a Drude contribution if appreciable at all should occur only at much lower frequencies than in the linear case. Note, we use $\lambda_{\text{s.o.}}$= 50 meV, which is the bulk value for Fe.

To check the accuracy of our electronic calculations we calculate first for Fe the frequency dependence of the linear polar Kerr angle $\phi_K^{(1)}(\omega)$=Re$\psi_K^{(1)}$ and compare with the experiment by Krinchik[12] and calculations by Oppeneer *et al.*[5] Results are shown in Fig. 1. Note, we find excellent agreement up to $\hbar\omega$ = 4 eV. The linear Kerr angle is of the order of 0.5°. The inclusion of the intraband contribution which changes $\phi_K^{(1)}(\omega)$ mainly for small ω leads to a further reduction of the linear Kerr angle and is responsible for changing $\phi_K^{(1)}$ to zero. It is of interest that our analysis reproduces also the enhancement of the linear Kerr angle at the plasma frequency as was already discussed by Feil and Haas.[13]

In Fig. 2 we show results for Fe in the case of the longitudinal Kerr geometry for the frequency dependence of the nonlinear Kerr angle $\phi_K^{(2)}(\omega)$=Re$\psi_K^{(2)}(\omega)$ for an angle of incidence of 45° and compare with the corresponding results for $\phi_K^{(1)}(\omega)$. We obtain for the optical range (up to 2.5 eV) a considerable enhancement of $\phi_K^{(2)}(\omega)$ over $\phi_K^{(1)}(\omega)$. For $\hbar\omega$ > 2.5 eV one has $|\phi_K^{(1)}(\omega)| \approx |\phi_K^{(2)}(\omega)| \approx$ 0.5° and for $\hbar\omega$ in the optical range one gets $\phi_K^{(1)}(\omega) \approx$ 0.5° while $\phi_K^{(2)}(\omega)$ may become 4° or larger. It is important to emphasize that the enhancement results largely from the term $\frac{\chi_1^{(2)}}{\chi_0^{(2)}}$ which has been neglected previously.[1] Note, the behavior of $\phi_K^{(1)}(\omega)$ and $\phi_K^{(2)}(\omega)$ for $\omega \to 0$ is controlled by the factor $\frac{\lambda_{\text{s.o.}}}{\hbar\omega}$.[11,14]

In Fig. 3 we demonstrate the enhancement of the Kerr rotation $\phi_K^{(2)}$ compared with the linear case as a function of the angle of incidence Θ_i for photon energies $\hbar\omega$ =0.6 eV and 3.6 eV. Figures 2 and 3 show that the enhancement of the nonlinear Kerr angle with respect to $\phi_K^{(1)}$ occurs over a large range of frequency and angle of incidence. Similar results are expected for a variety of magneto-optical Kerr geometries. The frequency dependence of the nonlinear Kerr angle reflects the surface electronic and magnetic structure and thus makes the NMOKE a material-sensitive probe.

It would be interesting to analyze the nonlinear Kerr angle and its symmetry properties for different magnetic

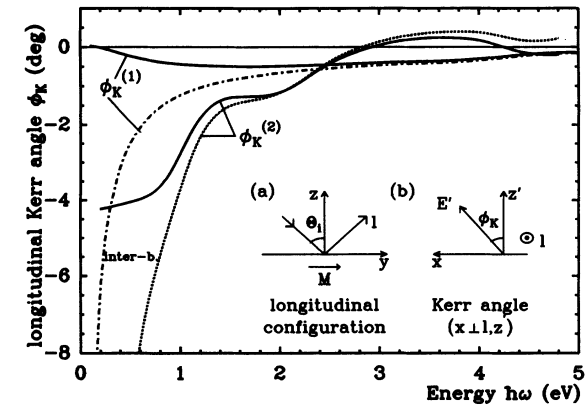

FIG. 2. Theoretical results for the frequency dependence of the nonlinear longitudinal Kerr angle $\phi_K^{(2)}(\omega)$ for Fe at an angle of incidence $\Theta_i = 45°$ using the same parameters. For comparison also our results for the linear longitudinal Kerr angle $\phi_K^{(1)}(\omega)$ are given. Insets (a) and (b) illustrate the definitions of the longitudinal configuration and the Kerr angle.

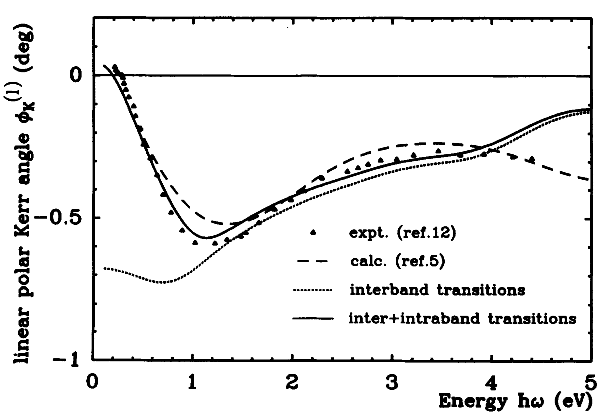

FIG. 1. Frequency dependence of the linear Kerr angle for the polar configuration. Theoretical results using parameters corresponding to Fe (see Ref. 11) are compared with experimental data. The interband contribution is shown separately.

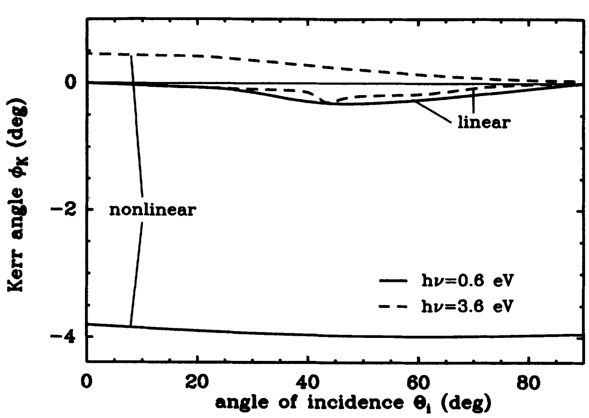

FIG. 3. Dependence of the Kerr angles $\phi_K^{(1)}$ and $\phi_K^{(2)}$ for Fe on the angle of incidence Θ_i. To demonstrate that $\phi_K^{(2)} > \phi_K^{(1)}$ we have chosen in view of the results shown in Fig. 2 the energies $\hbar\omega$ = 0.6 eV and 3.6 eV.

metals, semiconductors, insulators, clusters, and adsorbates. For materials with large spin-orbit coupling or large magnetic moments as in Heusler alloys particular large Kerr rotations could be expected.[3] Note, in materials where inversion symmetry is also absent in bulk, like in alloys, the NMOKE probes both surface and bulk. Magnetostriction changing the crystal symmetry and the magnetic easy axis may also effect the NMOKE. For thin magnetic films our theory would permit to determine surface magnetization, magnetic moments, and the magnetic anisotropy without involving hysteresis. Since inversion symmetry is broken at interfaces, these will contribute within the skin depth to the NMOKE. Our results imply that the nonlinear Kerr rotation is enhanced in all regions where inversion symmetry is broken. Here it may be of particular interest to investigate an interface of a magnetic and a nonmagnetic film with strong spin-orbit coupling.[15] It follows then from our theory that via hybridization the nonmagnetic material with its large $H_{\mathrm{s.o.}}$ contributes to the NMOKE.[11] For multilayers, the contributions of many interfaces may sum up coherently.

Finally, in order to give our results a more general significance we demonstrate in detail using Eq. (7) that nonlinear magneto-optics is a sensitive fingerprint of two-dimensional magnetism including magnetic anisotropy where the latter is considerably enhanced at the surface. Note, the basic quantities entering Eq. (7) are the magnetic susceptibility-tensor elements $\chi_1^{(j)}$. These are characterized by $(\chi^{(j)+} - \chi^{(j)-}) \sim \langle H_{\mathrm{s.o.}}\rangle\,|\mathbf{M}|$, where $\langle H_{\mathrm{s.o.}}\rangle \sim \lambda_{\mathrm{s.o.}}$, takes into account magnetic anisotropy and the effect of spin-orbit coupling on the electronic wave functions. Expanding the wave functions in the dipole matrix elements determining $\chi_1^{(j)}$ in terms of $\lambda_{\mathrm{s.o.}}$ one gets in leading order $F_{k1} \sim \chi_1^{(1)} \sim \lambda_{\mathrm{s.o.}}$, and $\chi_1^{(2)} \sim \lambda_{\mathrm{s.o.}}$.[11] It follows from this that $\tan\psi_K^{(2)} \sim \lambda_{\mathrm{s.o.}}$. The magnetization $\mathbf{M}$ enters linearly[16] and expresses the spin polarization of the electronic density of states. The *direction* of $\mathbf{M}$ given by an external magnetic field or by a magnetic easy axis will affect the tensor $\chi^{(j)}(\mathbf{M})$ only via $H_{\mathrm{s.o.}}$, the magnitude of which depends on magnetic anisotropy. SHG analyzed for different geometries reflects sensitively the fine structure (magnetic anisotropy) of surface magnetism. The nonlinear circular dichroic asymmetry[11] is given by $\Delta \sim \frac{I^+ - I^-}{I^+ + I^-} \sim (4\chi_1^{(2)}\chi_0^{(2)})/([\chi_0^{(2)}]^2 + [\chi_1^{(2)}]^2)$, where $I^{\pm}$ refers to the frequency-doubled light intensity of right- and left-hand circular polarization, and depends linearly on $|\mathbf{M}|$. Here, for an estimate we use $\chi_1^{(2)} \sim \langle H_{\mathrm{s.o.}}\rangle \int d\varepsilon [N_\uparrow(\varepsilon)N_\uparrow(\varepsilon + \hbar\omega)N_\uparrow(\varepsilon + 2\hbar\omega) - N_\downarrow(\varepsilon)N_\downarrow(\varepsilon + \hbar\omega)N_\downarrow(\varepsilon + 2\hbar\omega)]$.

Summarizing, we have shown that SHG is a sensitive fingerprint of magnetism in reduced dimensions. Due to the enhanced nonlinear surface polarization, magnetism affects much more strongly the light polarization and ellipticity of second-harmonic light than in linear optics. We have shown how to extend the Fresnel formulas to the nonlinear magnetic case. This is a result of general importance.

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