# Quantum theory of nonlinear and reciprocal properties of magneto-optical effects in paramagnetic media under a high magnetic field

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The nonlinear and reciprocal properties of magneto-optical (MO) effects in paramagnetic media under a high magnetic field are investigated by theoretical calculations. It is indicated that the Faraday effect in paramagnetic MO media has the obvious nonlinearity and reciprocity when the thermal energy $k_BT$ approaches the effective field energy $\mu_0\mu_BH_i$ ($\mu_0\mu_BH_e$). In addition, the MO effects, especially under an extremely high magnetic field, are found to depend essentially on the effective field $H_i$ except the magnetization $M$. Our results are shown to be consistent with the experimental data while they are dramatically contrary to the current widely-accepted conclusions of classical MO theories.

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## I. INTRODUCTION

Since the discovery of the Faraday effect in 1845, many magneto-optical (MO) effects, such as the Kerr, Voigt, and Zeeman effects, have been unveiled successively.[1]During the last decades, there has been considerable interest in the theoretical studies on MO effects.[2–5] To our knowledge, according to the classical MO theories, MO effects have the following basic properties: (1) Nonreciprocity, especially in the Faraday effect. In other words, the Faraday rotation $\theta$ should be the odd function of $M$ or $H_e$.[6–11] (2) Linear effects in paramagnetic media. Generally, $\theta = VH_eL$, where $V$ is the Verdet constant and $L$ the propagation length of light wave in media.[12–14] Actually most of the current MO devices, such as optical isolator and modulator, are fabricated on the basis of the nonreciprocal and linear properties of the MO effects. (3) The MO effects are essentially associated with $M$, rather than with $H_e$,[11,15,16] which manifests that the MO effects in strong magnetic media, for instance the ferromagnetic and ferrimagnetic media, are much stronger than those in paramagnetic and other weak magnetic media. That is, when the samples reach saturation magnetization, the MO effects in them, particularly the specific Faraday rotation $\theta_F$ ($\theta_F = \theta/L$), will not go on increasing with $H_e$.

In fact, these classical MO theories have greatly limited the region of research and applications of MO effects. Since Abulafya and Hansen *et al.* discovered weak nonlinear field-dependence of the specific Faraday rotation $\theta_F$ in yttrium iron garnet films (YIG) and other ferrimagnetic garnets in their experiments,[10,11,17] much attention has been paid to this field.[18] Recently, the nonlinear property of MO effects has been experimentally observed under a high magnetic field in paramagnetic media.[19–21] However, so far there are no corresponding theoretical interpretations of these experimental data. On the other hand, although a great deal of attention has been devoted to exposing the reciprocity in optics,[22] the detailed theory on the reciprocity in MO effects, especially the Faraday effect in paramagnetic media, has not been performed.

The purpose of the present paper is to theoretically study the nonlinear and reciprocal properties of the Faraday effect under a high magnetic field in paramagnetic media. Meanwhile, we successfully interpret the experimental phenomena in paramagnetic neodymium fluoride ($NdF_3$).[19] In addition, it is interesting to see that the MO effects are essentially related to the effective field $H_i$ rather than solely to $M$.

## II. THEORY

The MO property can be expressed in terms of the complex permittivity $\varepsilon$ tensor.[23–25] With the neglect of the local field acting on the electric dipole, the dielectric constant tensor elements can be written as

$$\varepsilon_{ij} = \varepsilon_0(\delta_{ij} + N\alpha_{ij}), \tag{1}$$

where $\alpha_{ij}$ is the $ij$ component of polarizability $\alpha$, $\delta_{ij}$ is the Dirac function, and $N$ is the atomic or ionic number per unit volume.

The components of polarizability are given by Condon and Shortley as[26]

$$\alpha_{ij} = \sum_a \frac{\rho_a}{\hbar} \sum_b \left[ \frac{P^i_{ab}P^j_{ba}}{\omega_0 + \omega - i\Gamma_{ab}} + \frac{P^j_{ab}P^i_{ba}}{\omega_0 - \omega + i\Gamma_{ab}} \right], \tag{2}$$

where $i, j = x, y, z$, and $\rho_a$ denotes the probability of an electron lying in the energy level $a$, and $\rho_{\max} = 1$, $P^x_{ab} = \langle \psi_a | ex | \psi_b \rangle$ is the element of electric dipole matrix. In the theoretical model, the ground state and the excited state are labeled by $a$ and $b$, respectively, $\omega$ represents the frequency of the incident light, $\hbar\omega_0$ is the energy-level separation between $a$ and $b$, and $\Gamma_{ab}$ is the linewidth.

For crystal systems whose symmetry is higher than that of orthorhombic crystal system, the specific Faraday rotation $\theta_F$ can be obtained from Eq. (2),

$$\theta_F = \frac{\omega_p^2\omega^2}{4nc} \sum_{a,b} \frac{\beta_a}{\omega_0} \cdot \frac{(\omega_0^2 - \omega^2 - \Gamma_{ab}^2)(f^+_{ab} - f^-_{ab})}{(\omega_0^2 - \omega^2 + \Gamma_{ab}^2)^2 + 4\omega^2\Gamma_{ab}^2}, \tag{3}$$

where $\omega_p^2 = \varepsilon_0 Ne^2/m$, and the oscillator strengths are

 

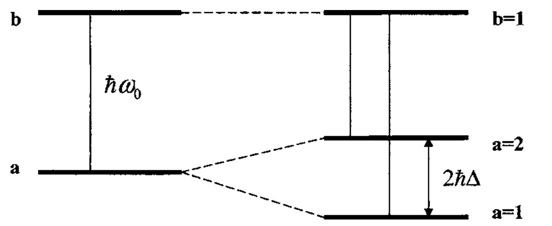


FIG. 1. The three-level model of the Faraday rotation in paramagnetic media.

$$f_{ab}^{\pm}=\frac{m\omega_{ab}}{\hbar e^2}|P_{ab}^{\pm}|^2. \tag{4}$$

At temperatures above $T_c$ ($T_N$), ferromagnetic, antiferromagnetic and ferrimagnetic media will change into paramagnetic media. In the paramagnetic media, owing to the exchange field $H_v$ and the applied field $H_e$, the ground level will be split as shown in Fig. 1.[27] As it is known, the effective exchange field $H_v$ in the paramagnetic media can be expressed as $vM$.[27,28] Here, $v$ is a coefficient related to the molecular field coefficient.[29] Therefore, the ground level splitting $\Delta E$ due to $H_v$ and $H_e$ is

$$\Delta E = 2\hbar\Delta = mg\mu_B\mu_0 H_i, \tag{5}$$

where $m$ is the magnetic quantum number, $g$ the Landé factor, $\mu_0$ the magnetic permeability in vacuum, $\mu_B$ the Bohr magneton, and the effective field $H_i$ is equal to $(H_e+vM)$.

The occupation probabilities of the two split ground states can be expressed as

$$\rho_{a1}=e^{-\beta E_1}\Big/\sum_l e^{-\beta E_l},$$

$$\rho_{a2}=e^{-\beta E_2}\Big/\sum_l e^{-\beta E_l}=\rho_{a1}e^{-2\beta\hbar\Delta}, \tag{6}$$

where $E_2=E_1+2\hbar\Delta$, $\beta=1/k_BT$, and $k_B$ is the Boltzmann constant. Suppose that

$$\Gamma_{11}=\Gamma_{21}=\Gamma, \quad \begin{matrix} f_{11}^{+}=0 \\ f_{11}^{-}=f^{-}\end{matrix}, \quad \begin{matrix} f_{21}^{+}=f^{+} \\ f_{21}^{-}=0\end{matrix}, \tag{7}$$

the specific Faraday rotation can be rewritten in another form

$$\theta_F=\frac{\omega_p^2\omega^2}{4nc}\left[-\frac{\rho_{a1}}{\omega_{11}}\cdot\frac{(\omega_{11}^2-\omega^2)f^-}{(\omega_{11}^2-\omega^2)^2+4\omega^2\Gamma^2}+\frac{\rho_{a2}}{\omega_{21}}\cdot\frac{(\omega_{21}^2-\omega^2)f^+}{(\omega_{21}^2-\omega^2)^2+4\omega^2\Gamma^2}\right]. \tag{8}$$

From Eq. (8), we can see that there will be electron distributions on the two split levels of the ground state when $H_i$ or $H_e$ are not extremely strong, i.e., $\rho_{a1},\rho_{a2}\neq 0$. Assume that the frequency shift caused by the ground level splitting is defined as

$$\omega_{11}=\omega_0+\Delta, \quad \omega_{21}=\omega_0-\Delta. \tag{9}$$

On the condition that $\Delta \ll \Gamma \ll \omega_0$, neglecting $\Gamma^2$ and $\Delta^2$, we then have

$$\theta_F=\frac{\omega_p^2\omega^2}{4n\omega_0^2c[(\omega_0^2-\omega^2)^2+4\omega^2\Gamma^2]}\times\left\{\begin{matrix}-\rho_{a1}(\omega_0^2+2\omega_0\Delta-\omega^2)(\omega_0-\Delta)f^- \\ +\rho_{a2}(\omega_0^2-2\omega_0\Delta-\omega^2)(\omega_0+\Delta)f^+\end{matrix}\right\}. \tag{10}$$

According to Eq. (6), $\rho_{a2}$ can be expanded as

$$\rho_{a2}=\rho_{a1}\left[1-2\beta\hbar\Delta+\frac{1}{2!}(2\beta\hbar\Delta)^2-\frac{1}{3!}(2\beta\hbar\Delta)^3+\cdots\right]. \tag{11}$$

As $M=\chi H_e$, $\chi$ is the magnetic susceptibility. Then $H_i$ can be expressed as

$$H_i=(1+v\chi)H_e. \tag{12}$$

Substituting Eqs. (5), (11), and (12) into Eq. (10), we can obtain

$$\theta_F=A_0+A_1H_e+A_2H_e^2+A_3H_e^3+A_4H_e^4+\cdots. \tag{13}$$

Define $\mathrm{Re}=\omega_p^2\omega^2\rho_{a1}/4nc\omega_0^2[(\omega_0^2-\omega^2)^2+4\omega^2\Gamma^2]$, thus

$$A_0=(\omega_0^2-\omega^2)\omega_0(f^+-f^-)\mathrm{Re};$$

$$A_1=-\frac{mg\mu_0\mu_B}{2\hbar}\left[(\omega_0^2+\omega^2)(f^++f^-)+\frac{2\hbar}{k_BT}(\omega_0^2-\omega^2)\omega_0f^+\right]\times(1+v\chi)\mathrm{Re};$$

$$A_2=\left(\frac{mg\mu_0\mu_B}{2\hbar}\right)^2\left[(\omega_0^2+\omega^2)+\frac{\hbar}{k_BT}(\omega_0^2-\omega^2)\omega_0\right]\times\frac{2\hbar}{k_BT}f^+(1+v\chi)^2\mathrm{Re};$$

$$A_3=-\left(\frac{mg\mu_0\mu_B}{2\hbar}\right)^3\left[\frac{1}{2}(\omega_0^2+\omega^2)+\frac{\hbar}{3k_BT}(\omega_0^2-\omega^2)\omega_0\right]\times\left(\frac{2\hbar}{k_BT}\right)^2f^+(1+v\chi)^3\mathrm{Re};$$

$$A_4=\left(\frac{mg\mu_0\mu_B}{2\hbar}\right)^4\left[\frac{1}{6}(\omega_0^2+\omega^2)+\frac{\hbar}{12k_BT}(\omega_0^2-\omega^2)\omega_0\right]\times\left(\frac{2\hbar}{k_BT}\right)^3f^+(1+v\chi)^4\mathrm{Re}. \tag{14}$$

Also, we can study the special case of $\rho_{a1}\to 1$, $\rho_{a2}\to 0$ in Eq. (8), in other words, nearly all the electrons occupy the lowest energy level where $H_i$ or $H_e$ are extremely strong. Then we have

$$\theta_F=-\frac{\omega_p^2\omega^2}{4nc}\cdot\frac{(\omega_{11}^2-\omega^2)f^-}{\omega_{11}[(\omega_{11}^2-\omega^2)^2+4\omega^2\Gamma^2]}. \tag{15}$$

From Eqs. (9), (12), and (15), $\theta_F$ can be written as

$$\theta_F = -\frac{\omega_p^2\omega^2 f^-}{4nc\omega_0^2[(\omega_0^2-\omega^2)^2+4\omega^2\Gamma^2]}\cdot\left[(\omega_0^2-\omega^2)\omega_0 - \frac{mg\mu_0\mu_B(\omega_0^2+\omega^2)}{2\hbar}\cdot H_i\right] = C_0 + C_1(1+v\chi)H_e, \tag{16}$$

where

$$C_0 = -\frac{\omega_p^2\omega^2 f^-}{4nc\omega_0^2[(\omega_0^2-\omega^2)^2+4\omega^2\Gamma^2]}(\omega_0^2-\omega^2)\omega_0,$$

$$C_1 = \frac{mg\mu_0\mu_B\omega_p^2\omega^2 f^-}{8nc\omega_0^2\hbar[(\omega_0^2-\omega^2)^2+4\omega^2\Gamma^2]}(\omega_0^2+\omega^2).$$

Here $\theta_F$ does not depend on temperature, but is linear with $H_i$ or $H_e$.

## III. RESULTS AND DISCUSSION

From Eq. (13), the following points should be noticed. (1) The specific Faraday rotation $\theta_F$ in paramagnetic media under a high magnetic field is nonlinear with $H_e$. (2) The existence of the even-order terms of $H_e$ will lead to the reciprocity in the Faraday effect.[30,22] (3) The magnitude of the reciprocal component is mainly related to $T$ and $H_e$, which implies that the reciprocity of the Faraday effect in paramagnetic media can not be neglected under a high magnetic field.

As for paramagnetic media, both the nonlinear and reciprocal properties are novel properties of the Faraday effect. From Eqs. (13) and (14), one can see that the relationship between the thermal energy $k_BT$ and the effective energy $\mu_0\mu_BH_i(\mu_0\mu_BH_e)$ is of great importance to the two properties. Thus, we can study the following three cases:

(i) $k_BT \gg \mu_0\mu_BH_i, \mu_0\mu_BH_e$. Under this condition, we can find $\rho_{a1} \cong \rho_{a2} \cong 1/2$, and the high-order terms of $H_i$ or $H_e$ in Eq. (13) tend to zero, which makes it hard for the nonlinearity and reciprocity in MO effects to be detected. The inequality will be satisfied in general magnetic field and room temperature, meanwhile, this fact explains why the above two MO properties are difficult to be observed in experiments under general conditions.

(ii) $k_BT \ll \mu_0\mu_BH_i, \mu_0\mu_BH_e$. This can be easily satisfied when $H_e$ ($H_i$) is extremely strong or $T$ is extremely low. Similarly, it is nearly the same as $\rho_{a2} \to 0$. Then from Eq. (16), we can see that, under the extreme conditions, the nonlinearity and reciprocity will not exist in the Faraday effect.

(iii) $k_BT \sim \mu_0\mu_BH_i, \mu_0\mu_BH_e$. It means that the value of $\mu_0\mu_BH_e(\mu_0\mu_BH_i)$ is close to that of the thermal energy $k_BT$. In other words, it corresponds to the situation of high effective field $H_i$ and low temperature $T$. In this case, the nonlinear and reciprocal properties of the MO effects will be very remarkable.

It is known that, according to the relationship $k_BT \sim \mu_0\mu_BH_i, \mu_0\mu_BH_e$, $T$ is about 18 K when $H_e \sim 200$ kOe. Therefore, when $H_e$ is up to 200 kOe, the nonlinear and reciprocal properties of the MO effects can be easily examined at about 18 K. Correspondingly, the Faraday rotation linearly depends on the applied magnetic field when the temperature is above 30 K in paramagnetic $NdF_3$, however, a strong nonlinearity is found below 30 K.[19] By means of the above theory, the nonlinear field-dependence of the Faraday effect at the 633 nm wavelength and low temperatures (<30 K) in $NdF_3$ is discussed. Fitting the above experimental results with Eq. (13), an excellent agreement is reached. However, in terms of the classical theory, provided that only the odd-order terms of $H_e$ are considered, the fitting results become much worse as displayed in Fig. 2(a). Different from Ref. 19, four terms of the expansion in Eq. (13) are taken into account. Obviously, it is easy to find that the coefficients of the odd-order terms ($A_1$, $A_3$) obtained from our analysis are negative while those of the even-order terms ($A_2$, $A_4$) are positive. Therefore, the even-order terms can not be neglected since they will greatly affect the value of the Faraday

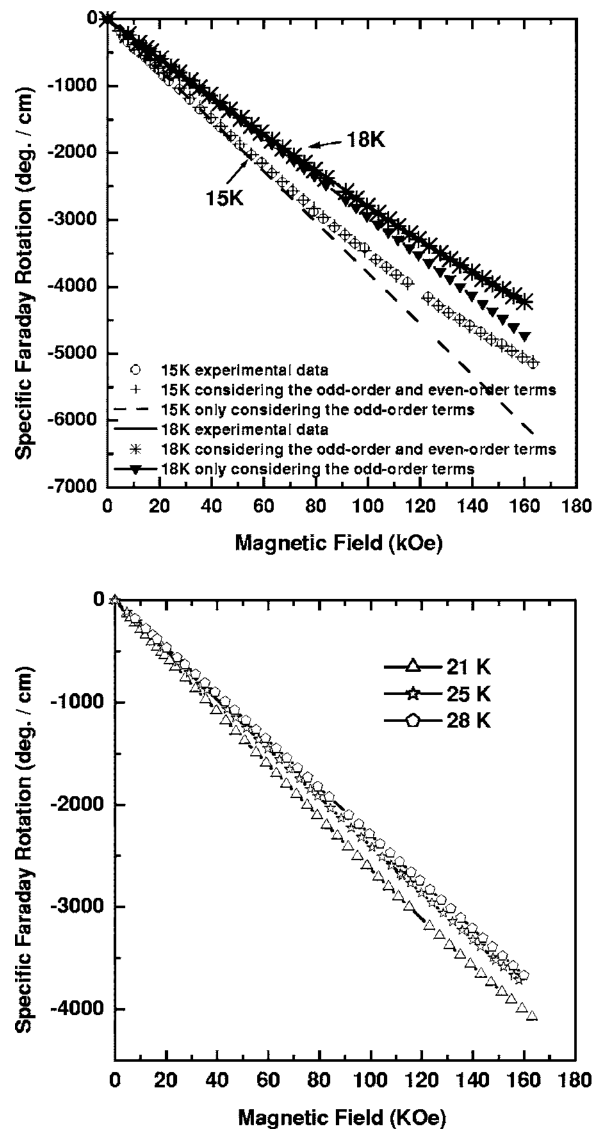


FIG. 2. (a) The comparison of $\theta_F$ vs $H_e$ between the theoretical results and the experiments at 15 K and 18 K in $NdF_3$. (b) Variation of the specific the Faraday rotation vs applied magnetic field at 21 K, 25 K, and 28 K in $NdF_3$.

TABLE I. Values of the coefficients ($v$, $A_1$, $A_2$, $A_3$, $A_4$) at different temperatures.

| | $v$ | $A_1$ | $A_2$ | $A_3$ | $A_4$ |
|---|---|---|---|---|---|
| 15 K | 865.3 | −40.26008 | 0.07589 | −3.70E−4 | 1.3878E−6 |
| 18 K | 779.9 | −31.30861 | 0.05252 | −3.30E−4 | 1.2282E−6 |
| 21 K | 716.3 | −28.21732 | 0.03142 | −2.06E−4 | 8.3044E−7 |
| 25 K | 682.8 | −24.53679 | 0.00644 | −4.00E−5 | 2.5493E−7 |
| 28 K | 625.4 | −22.91641 | 0.00023 | −9.68E−6 | 4.3368E−8 |

rotation. Thus, we point out that the Faraday effect shows an evident reciprocal property in $NdF_3$.

As for $NdF_3$, the electronic transition occurs mainly between the electron configurations $4f^3$ and $4f^25d$ of the trivalent neodymium ion. Based on the works of Diek[31] and Kramer,[32] the average energy of the $4f^25d$ configuration is selected as 43 000 $cm^{-1}$, and that of the ground configuration is zero, accordingly, $\hbar\omega_0 = 43\,000\ cm^{-1}$. The average refractive index $n$ of $NdF_3$ is taken as 1.6.[33] As there is no spontaneous Faraday rotation, the transition probabilities, following the work of Dionne *et al.*,[34,35] are described as $f^+ = f^- = f/2 = (m\omega_0/2h)(\langle\psi_a|x|\psi_b\rangle)^2$, where $x$ is the electric dipole operator. In calculating the Faraday rotation in $NdF_3$, the appropriate values of parameters $\Gamma$ and $f$ are found to be 4032.7 $cm^{-1}$ and $3.6896\times10^8$, respectively, through fitting the experimental data. Furthermore, the magnetic susceptibility $\chi$ conforms to the Curie-Weiss Law, where the Curie constant ($C$) is $1.58\pm0.01$ emu $mole^{-1}$ and the paramagnetic Curie temperature ($\Theta_P$) is $-19.3\pm1.8$ K.[21] Values of $v$ and the related coefficients ($A_1$, $A_2$, $A_3$, $A_4$) are listed in Table I.

Meanwhile, we present the variation of the Faraday rotation in $NdF_3$ with the applied field at different temperatures below 30 K in Figs. 2(a) and 2(b).[19] From the above figures and table, the following conclusions can be drawn: (1) $\theta_F$ will gradually be linear with $H_e$ when the temperature is up to 30 K. (2) In the media, with the increase of the temperature, the exchange interaction may be weakened or enhanced determined by the characteristic of the magnetic ions and the ion-next-nearest-neighbor distance, i.e., $v$ is dependent on the temperature $T$.[29]

Also, analyzing the above theoretical results, it is seen that nonlinear MO devices can be developed on the basis of the nonlinear property in MO media under a suitable applied field among proper temperature range. On the other hand, as it is known, the nonreciprocal MO devices applied in general applied fields and normal temperature range have been greatly developed. Nevertheless, because the reciprocal component may greatly affect the improvement of the isolation of MO devices, such as isolator, etc., it is necessary to take account of the MO reciprocity in designing MO devices.

Under extremely high applied field $H_e$, $M$ in the media can hardly increase with $H_e$, and thus, $H_e \gg vM$, however, in this instance, $\theta_F$ will still continue to increase with $H_e$ (or $H_i$), which suggests that the Faraday effect, especially when $H_e$ is extremely high, is primarily related to the applied field $H_e$ (or $H_i$) rather than solely to $M$. To describe it clearly, the fittings of the results in Fig. 2 are extended to the case of extremely high field as exhibited in Fig. 3. This figure indicates that $\theta_F$ in paramagnetic $NdF_3$ under extremely high field ($H_e > 600$ kOe) rises up to $10^5$ deg/cm which is much larger than that of saturated ferrimagnetic media, such as Bi-doped YIG under the general magnetic field. Therefore, it can be concluded that the Faraday effect is primarily determined by the effective field $H_i$. This opinion implies that in extremely high field and at low temperatures, even the weak magnetic media, such as paramagnetic media, etc., may present noticeable MO effects.

## IV. CONCLUSION

In this work, a theoretical investigation on MO effects in paramagnetic media under high magnetic field is presented. Based on the theory, the nonlinear relation between $\theta_F$ and $H_e$ or $H_i$ is given. In terms of the calculation, it is indicated that the reciprocal component can not be neglected under high magnetic field, which is different from the classical MO theory. Furthermore, an excellent agreement with the experimental results of $\theta_F$ vs $H_e$ at different temperatures below 30 K in paramagnetic $NdF_3$ is obtained. Finally, it is found that the MO effects, especially under extremely high magnetic field, are primarily related to the effective field $H_i$, rather than only to the magnetization $M$. Our results thus suggest that remarkable MO effects, such as much larger Faraday rotation, may be found in weak magnetic media under appropriate conditions.

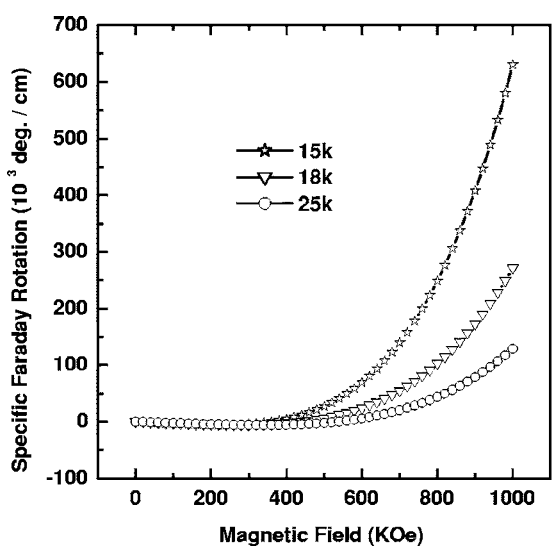


FIG. 3. $\theta_F$ vs $H_e$ in $NdF_3$ under extremely high field at 15 K, 18 K, and 25 K, respectively.

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